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(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Triazacyclohexane Derivatives

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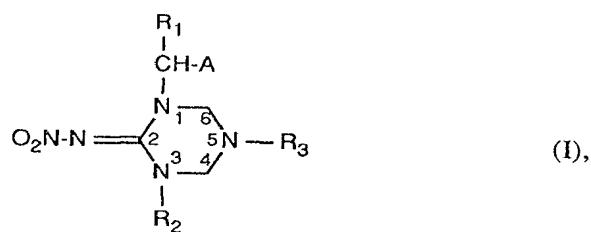
Notice: The specification contained herein as filed

Canada

CCA 3254 (10 89) 4*

~~2052731~~PI/5-18281/ATriazacyclohexane derivativesAbstract

Novel triazacyclohexane derivatives of formula I



wherein

 R_1 is hydrogen or $\text{C}_1\text{-}\text{C}_4$ alkyl; R_2 is hydrogen, $\text{C}_1\text{-}\text{C}_6$ alkyl, $\text{C}_3\text{-}\text{C}_6$ cycloalkyl or a radical $-\text{CH}_2\text{B}$; R_3 is hydrogen; $\text{C}_1\text{-}\text{C}_{10}$ alkyl; $\text{C}_3\text{-}\text{C}_6$ cycloalkyl; $\text{C}_1\text{-}\text{C}_{10}$ alkyl substituted by from 1 to 12 radicals from the group halogen, hydroxy, $\text{C}_1\text{-}\text{C}_4$ alkoxy, $\text{C}_1\text{-}\text{C}_4$ haloalkoxy havingfrom 1 to 9 halogen atoms, di-($\text{C}_1\text{-}\text{C}_4$ alkyl)amino and $\text{C}_1\text{-}\text{C}_5$ alkoxycarbonyl; $\text{C}_3\text{-}\text{C}_6$ cycloalkyl substituted by from 1 to 4 $\text{C}_1\text{-}\text{C}_4$ alkyl radicals or halogen atoms; $\text{C}_2\text{-}\text{C}_8$ alkenyl or $\text{C}_2\text{-}\text{C}_8$ alkynyl; $\text{C}_2\text{-}\text{C}_8$ alkenyl or $\text{C}_2\text{-}\text{C}_8$ alkynyl each of which is substituted by from 1 to 6 halogen atoms; phenyl; benzyl; or phenyl or benzyl each of which is substituted by from 1 to 3 ring substituents from the group halogen, $\text{C}_1\text{-}\text{C}_4$ alkyl, $\text{C}_1\text{-}\text{C}_4$ haloalkyl having from 1 to 9 halogen atoms, $\text{C}_1\text{-}\text{C}_4$ alkoxy, $\text{C}_1\text{-}\text{C}_4$ haloalkoxy having from 1 to 9 halogen atoms, $\text{C}_1\text{-}\text{C}_4$ alkylthio, nitro and cyano;

- A is an unsubstituted or mono- to tetra-substituted aromatic or non-aromatic, monocyclic or bicyclic heterocyclic radical that can have one or two substituents from the group $\text{C}_1\text{-}\text{C}_3$ haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl having from 1 to 3 halogen atoms, $\text{C}_2\text{-}\text{C}_3$ alkenyl, $\text{C}_2\text{-}\text{C}_3$ alkynyl, $\text{C}_2\text{-}\text{C}_3$ haloalkenyl and $\text{C}_2\text{-}\text{C}_3$ haloalkynyl each having from 1 to 4 halogen atoms, $\text{C}_1\text{-}\text{C}_3$ haloalkoxy having from 1 to 7 halogen atoms, $\text{C}_1\text{-}\text{C}_3$ alkylthio,

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$C_1\text{-}C_3$ haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, and from one to four substituents from the group $C_1\text{-}C_3$ alkyl, $C_1\text{-}C_3$ alkoxy and halogen; and

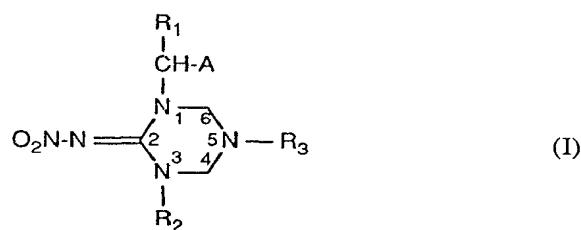
- B is phenyl; cyanophenyl; nitrophenyl; halophenyl having from 1 to 3 halogen atoms; phenyl substituted by $C_1\text{-}C_3$ alkyl, $C_1\text{-}C_3$ haloalkyl having from 1 to 7 halogen atoms, $C_1\text{-}C_3$ alkoxy or by $C_1\text{-}C_3$ haloalkoxy having from 1 to 7 halogen atoms; 3-pyridyl; 5-thiazolyl; 5-thiazolyl substituted by one or two substituents from the group $C_1\text{-}C_3$ -alkyl, $C_1\text{-}C_3$ haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, $C_2\text{-}C_3$ alkenyl, $C_2\text{-}C_3$ alkynyl, $C_1\text{-}C_3$ alkoxy, $C_2\text{-}C_3$ haloalkenyl and $C_2\text{-}C_3$ haloalkynyl each having from 1 to 4 halogen atoms, $C_1\text{-}C_3$ haloalkoxy having from 1 to 7 halogen atoms, $C_1\text{-}C_3$ alkylthio, $C_1\text{-}C_3$ haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, halogen, cyano and nitro; or 3-pyridyl substituted by one or two radicals from the group $C_1\text{-}C_3$ haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, $C_2\text{-}C_3$ alkenyl, $C_2\text{-}C_3$ alkynyl, $C_2\text{-}C_3$ haloalkenyl and $C_2\text{-}C_3$ -haloalkynyl each having from 1 to 4 halogen atoms, $C_1\text{-}C_3$ haloalkoxy having from 1 to 7 halogen atoms, $C_1\text{-}C_3$ alkylthio, $C_1\text{-}C_3$ haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, or by from one to four radicals from the group $C_1\text{-}C_3$ alkyl, $C_1\text{-}C_3$ -alkoxy and halogen;

and salts thereof with inorganic acids, have valuable pesticidal properties. Compositions comprising those compounds, processes for the preparation thereof, and their use as pesticides, especially as insecticides and acaricides in agriculture, are described.

Triazacyclohexane derivatives

The present invention relates to novel substituted 2-nitroimino-1,3,5-triazacyclohexane derivatives, to processes for the preparation thereof, to pesticides that comprise those compounds, and to their use in the control of pests.

The triazacyclohexane derivatives according to the invention correspond to formula I



wherein

R₁ is hydrogen or C₁-C₄alkyl;

R₂ is hydrogen, C₁-C₆alkyl, C₃-C₆cycloalkyl or a radical -CH₂B;

R₃ is hydrogen; C₁-C₁₀alkyl; C₃-C₆cycloalkyl; C₁-C₁₀alkyl substituted by from 1 to 12 radicals from the group halogen, hydroxy, C₁-C₄alkoxy, C₁-C₄haloalkoxy having

from 1 to 9 halogen atoms, di-(C₁-C₄alkyl)amino and C₁-C₅alkoxycarbonyl;

C₃-C₆cycloalkyl substituted by from 1 to 4 C₁-C₄alkyl radicals or halogen atoms;

C₂-C₈alkenyl or C₂-C₈alkynyl; C₂-C₈alkenyl or C₂-C₈alkynyl each of which is substituted by from 1 to 6 halogen atoms; phenyl; benzyl; or phenyl or benzyl each of which is substituted by from 1 to 3 ring substituents from the group halogen,

C₁-C₄alkyl, C₁-C₄haloalkyl having from 1 to 9 halogen atoms, C₁-C₄alkoxy,

C₁-C₄haloalkoxy having from 1 to 9 halogen atoms, C₁-C₄alkylthio, nitro and cyano;

A is an unsubstituted or mono- to tetra-substituted aromatic or non-aromatic, monocyclic or bicyclic heterocyclic radical that can have one or two substituents from the group C₁-C₃haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl having from 1 to 3 halogen atoms, C₂-C₃alkenyl, C₂-C₃alkynyl,

C_2 - C_3 haloalkenyl and C_2 - C_3 haloalkynyl each having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, and from one to four substituents from the group C_1 - C_3 alkyl, C_1 - C_3 alkoxy and halogen; and

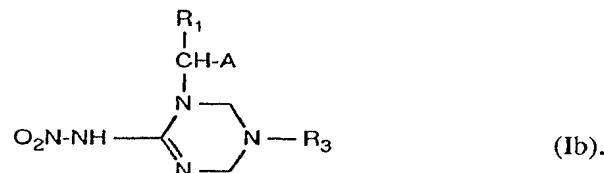
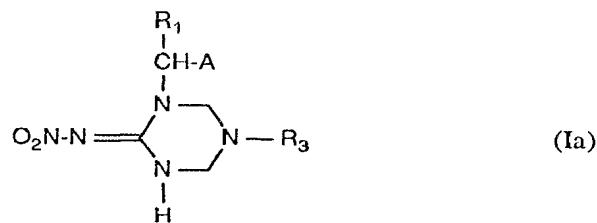
- B is phenyl; cyanophenyl; nitrophenyl; halophenyl having from 1 to 3 halogen atoms; phenyl substituted by C_1 - C_3 alkyl, C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, C_1 - C_3 alkoxy or by C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms; 3-pyridyl; 5-thiazolyl; 5-thiazolyl substituted by one or two substituents from the group C_1 - C_3 alkyl, C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, C_2 - C_3 alkenyl, C_2 - C_3 alkynyl, C_1 - C_3 alkoxy, C_2 - C_3 haloalkenyl and C_2 - C_3 haloalkynyl each having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, halogen, cyano and nitro; or 3-pyridyl substituted by one or two radicals from the group C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, C_2 - C_3 alkenyl, C_2 - C_3 alkynyl, C_2 - C_3 haloalkenyl and C_2 - C_3 -haloalkynyl each having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, or by from one to four radicals from the group C_1 - C_3 alkyl, C_1 - C_3 alkoxy and halogen;

and salts thereof with inorganic acids.

The compounds of formula I according to the invention also include the salts thereof with agrochemically tolerable inorganic acids. Examples of such acids are hydrochloric acid, hydrobromic acid, sulfuric acid, phosphoric acid and nitric acid, and also acids having the same central atom and higher or lower degrees of oxidation, such as perchloric acid, nitrous acid or phosphorous acid.

The compounds of formula I can occur in tautomeric forms Ia or Ib when the radical R_2 is hydrogen:

- 3 -



The compounds of formula I can also occur as double-bond isomers with respect to N=C(2).

Formula I according to the invention is therefore to be understood as including formulae Ia and Ib and the double-bond isomers.

In the definition of formula I according to the invention, the individual generic terms are to be understood as having the following meanings:

The halogen atoms that come into consideration as substituents are fluorine and chlorine and also bromine and iodine, with fluorine, chlorine and bromine being preferred. Halogen is here to be understood as being an independent substituent or part of a substituent, such as in haloalkyl, haloalkylthio, haloalkoxy, halocycloalkyl, haloalkenyl, haloalkynyl, haloallyloxy or haloallylthio. The alkyl, alkylthio, alkenyl, alkynyl and alkoxy radicals that come into consideration as substituents can be straight-chain or branched. There may be mentioned as examples of such alkyl radicals methyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec-butyl and tert-butyl. Suitable alkoxy radicals are *inter alia*: methoxy, ethoxy, propoxy, isopropoxy, or butoxy and its isomers. Alkylthio is, for example, methylthio, ethylthio, isopropylthio, propylthio or the isomers of butylthio. If the alkyl, alkoxy, alkenyl, alkynyl or cycloalkyl groups that come into consideration as substituents are substituted by halogen, they may be only partially halogenated or alternatively per-halogenated. Halogen, alkyl and alkoxy here have the definitions given above. Examples of the alkyl elements of those groups are methyl substituted from one to three

times by fluorine, chlorine and/or by bromine, for example CHF₂ or CF₃; ethyl substituted from one to five times by fluorine, chlorine and/or by bromine, for example CH₂CF₃, CF₂CF₃, CF₂CCl₃, CF₂CHCl₂, CF₂CHF₂, CF₂CFCI₂, CF₂CHBr₂, CF₂CHClF, CF₂CHBrF or CCIFCHClF; propyl or isopropyl substituted from one to seven times by fluorine, chlorine and/or by bromine, for example CH₂CHBrCH₂Br, CF₂CHFCF₃, CH₂CF₂CF₃ or CH(CF₃)₂; butyl or one of its isomers substituted from one to nine times by fluorine, chlorine and/or by bromine, for example CF(CF₃)CHFCF₃ or CH₂(CF₂)₂CF₃; 2-chlorocyclopropyl or 2,2-difluorocyclopropyl; 2,2-difluorovinyl, 2,2-dichlorovinyl, 2-chloroalkyl, 2,3-dichlorovinyl or 2,3-dibromovinyl.

If the alkyl, alkoxy or cycloalkyl groups defined are substituted by other substituents, they may be mono- or poly-substituted by identical or different substituents selected from those listed. Preferably, one or two other substituents are present in the substituted groups. The cycloalkyl radicals that come into consideration as substituents are, for example, cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl. Alkenyl and alkynyl groups contain one unsaturated carbon-carbon bond. Typical examples are allyl, methallyl or propargyl, but also vinyl and ethynyl. The double or triple bonds in allyloxy, propargyloxy, allylthio or propargylthio are separated from the point of linkage to the hetero atom (O or S) preferably by a saturated carbon atom.

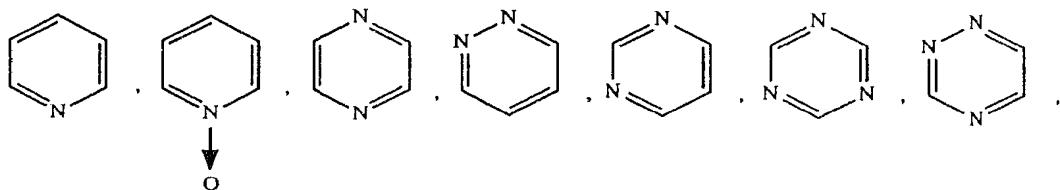
Of the compounds of formula I defined above, prominence is to be given to those wherein the radical

R₃ is C₅-C₁₀alkyl; C₃-C₆cycloalkyl; C₁-C₁₀alkyl substituted by from 1 to 12 radicals from the group halogen, hydroxy, C₁-C₄alkoxy, C₁-C₄haloalkoxy having from 1 to 9 halogen atoms, di-(C₁-C₄alkyl)amino and C₁-C₅alkoxycarbonyl; C₃-C₆cycloalkyl substituted by from 1 to 4 C₁-C₄alkyl radicals or halogen atoms; C₂-C₈alkenyl or C₂-C₈alkynyl; C₂-C₈alkenyl or C₂-C₈alkynyl each of which is substituted by from 1 to 6 halogen atoms; phenyl; benzyl; or phenyl or benzyl each of which is substituted by from 1 to 3 ring substituents from the group halogen, C₁-C₄alkyl, C₁-C₄haloalkyl having from 1 to 9 halogen atoms, C₁-C₄alkoxy, C₁-C₄haloalkoxy having from 1 to 9 halogen atoms, C₁-C₄alkylthio, nitro and cyano; and R₁, R₂ and A are as defined above.

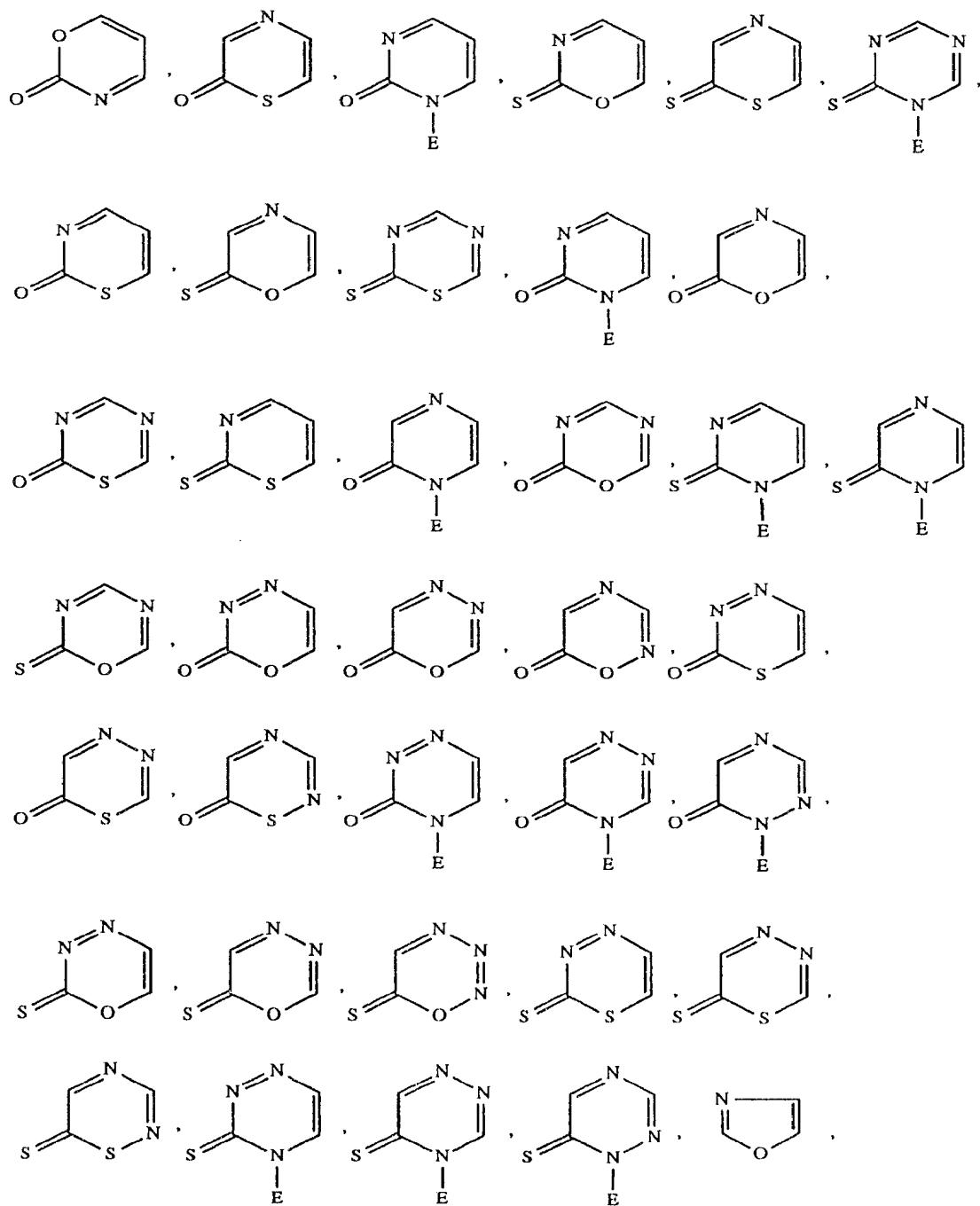
Of special importance according to the invention are also those compounds of formula I wherein the heterocyclic radical A is unsaturated, is bonded via a carbon atom to the radical of the molecule of the compound of formula I and contains at least one nitrogen

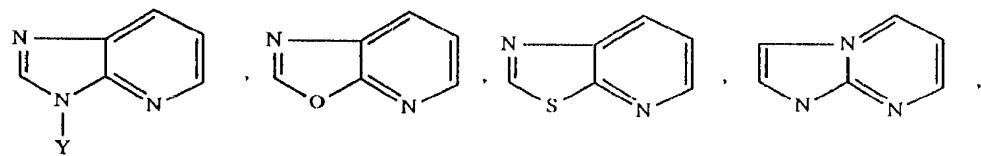
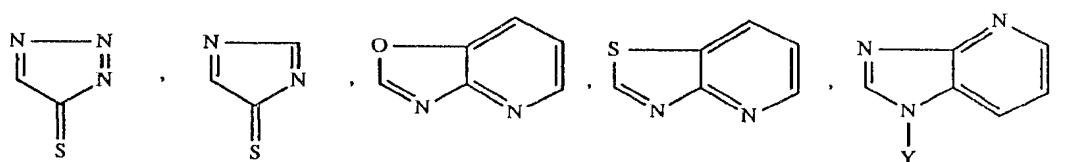
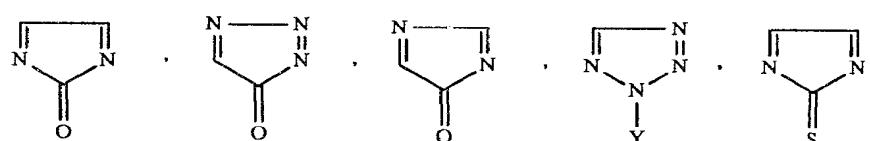
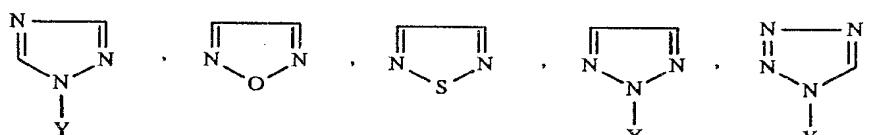
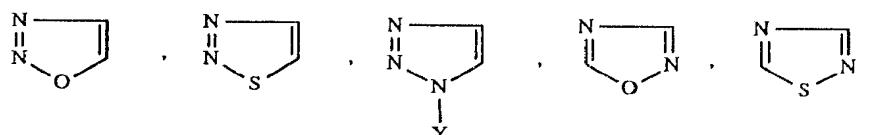
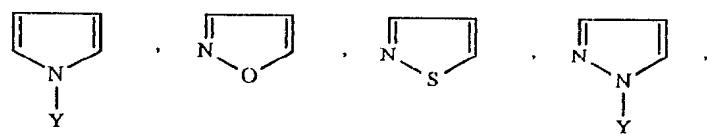
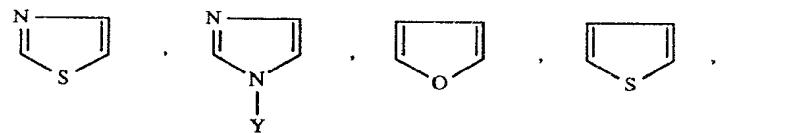
atom; those compounds wherein the heterocyclic radical A is unsaturated, is bonded via a carbon atom to the radical of the molecule of the compound of formula I and contains from one to three hetero atoms from the group oxygen, sulfur and nitrogen, not more than one oxygen or sulfur atom being present; and those compounds wherein the heterocyclic radical A contains from one to three hetero atoms from the group oxygen, sulfur and nitrogen, of which one hetero atom is always nitrogen, not more than one oxygen atom or sulfur atom being present.

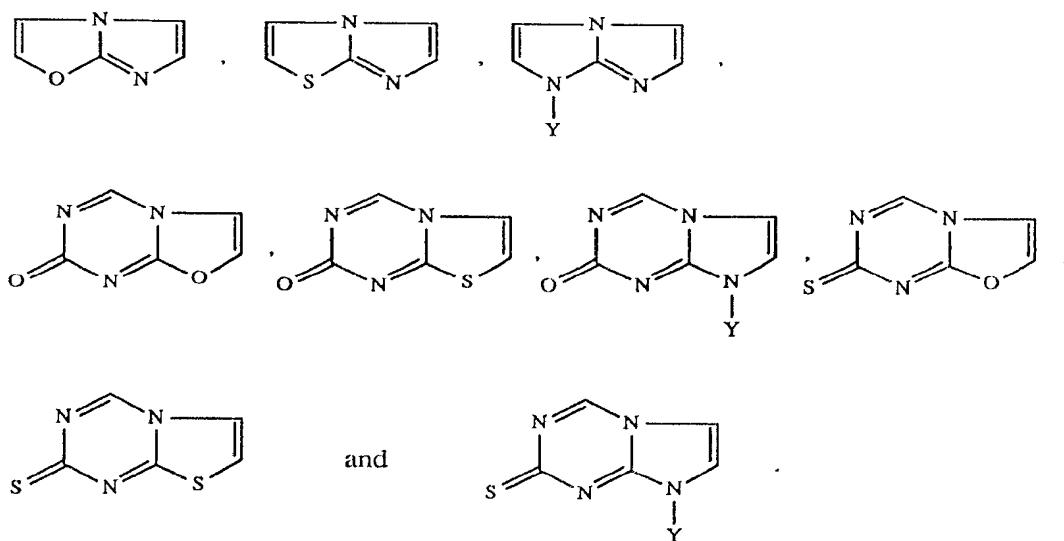
The ring systems covered by the definition of the heterocyclic radical A are significant as regards the biological activity of the compounds of formula I according to the invention. These ring systems contain at least one hetero atom as ring member, that is to say at least one of the atoms forming the basic cyclic structure is other than carbon. In principle, all atoms of the periodic system of the elements are capable of acting as ring members, provided they are able to form at least two covalent bonds. The heterocyclic radical is preferably unsaturated and bonded to the basic structure of formula I via a carbon atom as ring member. Unsaturated ring systems of the definition A contain one or more double bonds; such ring systems are preferably polyunsaturated and are generally of aromatic nature. Preference is given to ring systems that contain at least one nitrogen atom as hetero atom. Such rings of the definition A usually contain from one to three hetero atoms from the group oxygen, sulfur and nitrogen, not more than one oxygen or sulfur atom being present. Preference is given to ring systems of the definition of A wherein the heterocyclic radical A contains from one to three hetero atoms from the group oxygen, sulfur and nitrogen, of which one hetero atom is always nitrogen, not more than one oxygen atom or sulfur atom being present. Examples of heterocycles of definition A according to the invention are to be found especially in the following group of basic structures:



- 6 -







In the above formulae, E is C₁-C₃alkyl and Y is hydrogen, C₁-C₃alkyl or cyclopropyl.

The heterocycles A listed as examples above can be unsubstituted or, depending on the number of substituents possible in the ring system, can carry up to four of the substituents indicated under formula I. Preferably, these heterocycles carry from one to three substituents from the group halogen, C₁-C₃alkyl, C₁-C₃haloalkyl and C₁-C₃haloalkoxy each having from 1 to 7 halogen atoms, and C₁-C₃alkoxy. Especially preferred heterocycles A are pyridyl radicals or thiazolyl radicals, for example 3-pyridyl, 2-halopyrid-5-yl, 2,3-dihalopyrid-5-yl, 2-halothiazol-4-yl, 1-oxopyrid-3-yl, 1-oxo-2-halopyrid-5-yl and 1-oxo-2,3-dihalopyrid-5-yl.

In the compounds of formula I, the radical B is preferably a phenyl, pyridyl or thiazolyl radical that can be unsubstituted or substituted by one or two radicals from the group halogen, C₁-C₃alkyl, C₁-C₃haloalkyl and C₁-C₃haloalkoxy each having from 1 to 7 halogen atoms, and C₁-C₃alkoxy.

Of the compounds of formula I, prominence is to be given, on account of their biological properties, to those compounds wherein R₁ is hydrogen, R₂ is methyl, ethyl or cyclopropyl, and A is pyridyl, 1-oxopyridyl or thiazolyl, or is pyridyl, 1-oxopyridyl or thiazolyl each of which is substituted by from one to three substituents from the group halogen, C₁-C₃alkyl, C₁-C₃haloalkyl and C₁-C₃haloalkoxy each having from 1 to 7

halogen atoms, and C₁-C₃alkoxy. Within this meaning, also of interest are those compounds of formula I wherein

- a) R₁ is hydrogen; and/or
- b) R₂ is methyl; and/or
- c) R₃ is C₁-C₃alkyl, cyclopropyl, cyclohexyl, phenyl, benzyl or the radical -CH₂-COO-CH₃.

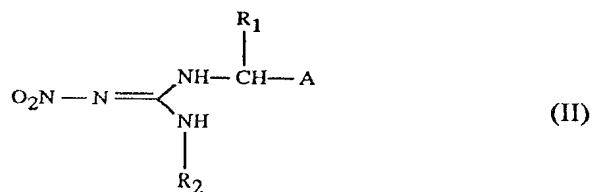
Also of interest in accordance with the invention are those classes of compound of formula I wherein

- R₃ is benzyl or phenyl each of which is substituted by from 1 to 3 ring substituents from the group fluorine, chlorine, bromine, C₁-C₂alkyl, C₁-C₂haloalkyl, C₁-C₂alkoxy, C₁-C₂alkylthio, nitro and cyano;
- R₃ is C₁-C₆alkyl substituted by a hydroxy group;
- R₃ is C₁-C₆alkyl substituted by a C₁-C₅alkoxycarbonyl group;
- R₃ is -CH₂CH₂F, -CH₂CH₂Br, -CH₂CH₂CH₂Cl, -CH₂CH₂CH₂Br or -CH₂CHClCH₂CH₂Cl;
- R₃ is -CH₂CH₂O-CH₃, -CH₂CH₂CH₂-O-CH₂CH₃, -CH(CH₃)CH₂-O-CH₃, -CH₂CH(OCH₃)₂, -CH₂CH₂-N(CH₂CH₃)₂, -CH₂-CH₂CH₂-N(CH₃)₂ or -CH₂CH₂CH₂-N(CH₂CH₃)₂;
- R₃ is C₄-C₆cycloalkyl that is unsubstituted or substituted by one or two C₁-C₄alkyl radicals;
- R₃ is cyclopentyl or cyclohexyl;
- R₃ is C₃-C₆cycloalkyl substituted by one or two methyl groups;
- A is 2-chlorothiazol-4-yl, 2,3-dichloropyrid-5-yl, 1-oxopyrid-3-yl or 1-oxo-2-chloropyrid-5-yl; R₂ is methyl and R₃ is cyclopropyl, -CH₂CH₂Cl, -CH₂CH(OCH₃)₂ or -CH₂CH₂N(CH₃)₂;
- A is 2-chlorothiazol-4-yl;
- A is 2-chloropyrid-5-yl; or
- A is 2-chloropyrid-5-yl, 2,3-dichloropyrid-5-yl, 2-chlorothiazol-4-yl, 1-oxopyrid-3-yl or 1-oxo-2-chloropyrid-5-yl; R₁ is hydrogen; R₂ is methyl; and R₃ is n-propyl.

The compounds of formula I according to the invention can be prepared by, for example,

- a) reacting a compound of formula II

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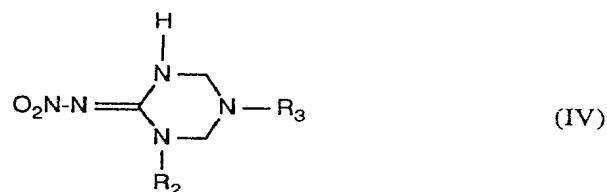


with formaldehyde, or paraformaldehyde, and a compound of formula III



or

- b) reacting a compound of formula IV



with a compound of formula V



or

- c) for the preparation of a compound of formula I wherein R₂ is other than hydrogen, reacting a resulting compound of formula I wherein R₂ is hydrogen with a compound of formula VI



and, if desired, converting a resulting compound of formula I into a salt thereof in a

manner known *per se*; R₁, R₂, R₃ and A in formulae II to VI being as defined above, X being halogen and Y being a leaving group. There may come into consideration as leaving groups X and Y, for example: halogen, preferably chlorine, bromine or iodine, or sulfonic acid radicals, such as alkanesulfonic acid radicals, mesylate or tosylate.

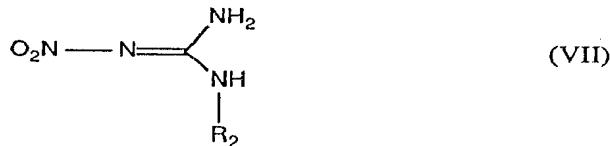
Variant a) of the above process according to the invention is advantageously carried out under normal pressure, but may also be carried out under elevated pressure, in an inert solvent and at temperatures of from 0°C to +140°C, especially from +20°C to +120°C. Suitable solvents are especially alcohols, such as methanol, ethanol and propanol, and also water. Other suitable solvents are, for example, aromatic hydrocarbons, such as benzene, toluene and xylene; ethers, such as tetrahydrofuran, dioxane and diethyl ether; halogenated hydrocarbons, such as methylene chloride, chloroform, carbon tetrachloride and chlorobenzene, and other solvents that do not impair the reaction. The solvents may also be used as mixtures. The reaction may be carried out with the addition of an acid catalyst, such as HCl, H₂SO₄, or a sulfonic acid, such as p-toluenesulfonic acid. The resulting water of reaction can, where necessary, be removed by means of a water separator or by the addition of a molecular sieve.

The above-mentioned process variants b) and c) can preferably be carried out under normal or slightly elevated pressure and in the presence of preferably aprotic solvents or diluents. Suitable solvents or diluents are, for example, ethers and ethereal compounds, such as diethyl ether, dipropyl ether, dibutyl ether, dioxane, dimethoxyethane and tetrahydrofuran; aliphatic, aromatic and halogenated hydrocarbons, especially benzene, toluene, xylene, chloroform, methylene chloride, carbon tetrachloride and chlorobenzene; nitriles, such as acetonitrile or propionitrile; dimethyl sulfoxide and dimethylformamide. The processes are generally carried out at a temperature of from -20 to +140°C, preferably from 0 to +120°C, preferably in the presence of a base. Examples of suitable bases are carbonates, such as sodium and potassium carbonate. Hydrides, such as sodium hydride, potassium hydride and calcium hydride, can also be used as bases.

The starting materials of formulae II, III, V and VI are known or can be prepared analogously to known processes.

The 2-nitroguanidine derivatives used as starting materials of formula II, and the preparation thereof, are known from EP Patent Applications 375 907 and 376 279. The primary amines of formula III are products that are readily available commercially.

The 2-nitroimino-1,3,5-triazoles of formula IV, to which the present invention also relates, are obtainable by reacting a 2-nitroguanidine of formula VII



with formaldehyde, or paraformaldehyde, and a compound of formula III



R_2 and R_3 in formulae VII and III being as defined above. The reaction conditions for this process are the same as those for process variant a) above for the preparation of the compounds of formula I. The compounds of formula IV are novel with the exception of 2-nitroimino-5-methyl-1,3,5-triazacyclohexane (EP Patent Application 0 386 565) and 2-nitroimino-1,3,5-triazacyclohexane (US-PS 4 937 340). The nitroguanidines of formula VII are known (see US-PS 4 804 780 and 4 221 802) or can be prepared in analogous manner.

A large number of compounds of formula V are known (see, for example, EP Patent Applications 375 907 and 376 279). There are preferred as starting materials those compounds of formula V wherein X is chlorine.

Similarly, a large number of compounds of formula VI are known. They are products that are commercially available or that are readily obtainable analogously to known processes. The leaving group Y in those compounds is preferably a halogen atom, especially chlorine.

It is already known that some open-chained 2-nitroguanidine derivatives have pesticidal properties (see, for example, EP Patent Applications 0 375 907 and 0 376 279). However, pesticidal heterocyclic compounds based on a nitroguanidine structure are also known. For example, EP Patent Applications 0 192 060 and 0 259 738 describe 2-nitroiminopyrimidine derivatives having insecticidal activity. Furthermore, in US-PS 4 937 340, 2-nitroimino-1,3,5-triazacyclohexane and other corresponding

derivatives containing nitro groups are proposed as additives for explosives. Insecticidal compounds of the type according to the invention are proposed in EP Patent Application 0 386 565, the compounds of formula I according to the invention being partially covered by the broad scope of the claims of this EP Patent Application.

Surprisingly, it has been found that the compounds of formula I according to the invention are valuable active ingredients in pest control while being well tolerated by warm-blooded animals, fish and plants. The compounds according to the invention can be used especially against insects that occur on useful plants and ornamentals in agriculture, especially in cotton, vegetable and fruit crops, in forestry, in the protection of stored goods and material stocks, and also in the hygiene sector, especially on domestic animals and productive livestock. The compounds are effective especially against plant-destructive sucking insects, especially against aphids and cicadas. They are effective against all or individual development stages of normally sensitive and also resistant species. Their action may manifest itself in the death of the pests immediately or only at a later date, for example at moulting, or in reduced oviposition and/or a reduced hatching rate. The above-mentioned pests include:

of the order Lepidoptera, for example

Acleris spp., Adoxophyes spp., Aegeria spp., Agrotis spp., Alabama argillaceae, Amylois spp., Anticarsia gemmatalis, Archips spp., Argyrotaenia spp., Autographa spp., Busseola fusca, Cadra cautella, Carposina nipponensis, Chilo spp., Choristoneura spp., Clysia ambiguella, Cnaphalocrocis spp., Cnephasia spp., Cochylis spp., Coleophora spp., Crocidolomia binotalis, Cryptophlebia leucotreta, Cydia spp., Diatraea spp., Diparopsis castanea, Earias spp., Ephestia spp., Eucosma spp., Eupoecilia ambiguella, Euproctis spp., Euxoa spp., Grapholita spp., Hedyia nubiferana, Heliothis spp., Hellula undalis, Hyphantria cunea, Keiferia lycopersicella, Leucoptera scitella, Lithocollethis spp., Lobesia botrana, Lymantria spp., Lyonetia spp., Malacosoma spp., Mamestra brassicae, Manduca sexta, Operophtera spp., Ostrinia nubilalis, Pammene spp., Pandemis spp., Panolis flammea, Pectinophora gossypiella, Phthorimaea operculella, Pieris rapae, Pieris spp., Plutella xylostella, Prays spp., Scirpophaga spp., Sesamia spp., Sparganothis spp., Spodoptera spp., Synanthedon spp., Thaumetopoea spp., Tortrix spp., Trichoplusia ni and Yponomeuta spp.;

of the order Coleoptera, for example

Agriotes spp., Anthonomus spp., Atomaria linearis, Chaetocnema tibialis, Cosmopolites spp., Curculio spp., Dermestes spp., Diabrotica spp., Epilachna spp., Eremnus spp., Leptinotarsa decemlineata, Lissorhoptrus spp., Melolontha spp., Orycaophilus spp.,

Otiorhynchus spp., Phlyctinus spp., Popillia spp., Psylliodes spp., Rhizopertha spp., Scarabaeidae, Sitophilus spp., Sitotroga spp., Tenebrio spp., Tribolium spp. and Trogoderma spp.; of the order Orthoptera, for example Blatta spp., Blattella spp., Gryllotalpa spp., Leucophaea maderae, Locusta spp., Periplaneta spp. and Schistocerca spp.; of the order Isoptera, for example

Reticulitermes spp.; of the order Psocoptera, for example Liposcelis spp.; of the order Anoplura, for example Haematopinus spp., Linognathus spp., Pediculus spp., Pemphigus spp. and Phylloxera spp.; of the order Mallophaga, for example Damalinea spp. and Trichodectes spp.;

of the order Thysanoptera, for example

Frankliniella spp., Hercinothrips spp., Taeniothrips spp., Thrips palmi, Thrips tabaci and Scirtothrips aurantii;

of the order Heteroptera, for example

Cimex spp., Distantiella theobroma, Dysdercus spp., Euchistus spp., Eurygaster spp., Leptocorisa spp., Nezara spp., Piesma spp., Rhodnius spp., Sahlbergella singularis, Scotinophara spp. and Triatoma spp.;

of the order Homoptera, for example

Aleurothrixus floccosus, Aleyrodes brassicae, Aonidiella spp., Aphididae, Aphis spp., Aspidiotus spp., Bemisia tabaci, Ceroplaste spp., Chrysomphalus aonidium, Chrysomphalus dictyospermi, Coccus hesperidum, Empoasca spp., Eriosoma larigerum, Erythroneura spp., Gascardia spp., Laodelphax spp., Lecanium corni, Lepidosaphes spp., Macrosiphus spp., Myzus spp., Nephrotettix spp., Nilaparvata spp., Paratoria spp., Pemphigus spp., Planococcus spp., Pseudaulacaspis spp., Pseudococcus spp., Psylla spp., Pulvinaria aethiopica, Quadraspidiotus spp., Rhopalosiphum spp., Saissetia spp., Scaphoideus spp., Schizaphis spp., Sitobion spp., Trialeurodes vaporariorum, Trioza erytreae and Unaspis citri;

of the order Hymenoptera, for example

Acromyrmex, Atta spp., Cephus spp., Diprion spp., Diprionidae, Gilpinia polytoma, Hoplocampa spp., Lasius spp., Monomorium pharaonis, Neodiprion spp., Solenopsis spp. and Vespa spp.;

of the order Diptera, for example

Aedes spp., Antherigona soccata, Bibio hortulanus, Calliphora erythrocephala, Ceratitis spp., Chrysomyia spp., Culex spp., Cuterebra spp., Dacus spp., Drosophila melanogaster, Fannia spp., Gastrophilus spp., Glossina spp., Hypoderma spp., Hyppobosca spp., Liriomyza spp., Lucilia spp., Melanagromyza spp., Musca spp., Oestrus spp., Orseolia spp., Oscinella frit, Pegomyia hyoscyami, Phobia spp., Rhagoletis pomonella, Sciara spp.,

Stomoxys spp., Tabanus spp., Tannia spp. and Tipula spp.;
of the order Siphonaptera, for example
Ceratophyllus spp., Xenopsylla cheopis;
of the order Acarina, for example
Acarus siro, Aceria sheldoni, Aculus schlechtendali, Amblyomma spp., Argas spp.,
Boophilus spp., Brevipalpus spp., Bryobia praetiosa, Calipitrimerus spp., Chorioptes spp.,
Dermanyssus gallinae, Eotetranychus carpini, Eriophyes spp., Hyalomma spp., Ixodes
spp., Oligonychus pratensis, Ornithodoros spp., Panonychus spp., Phyllocoptuta oleivora,
Polyphagotarsonemus latus, Psoroptes spp., Rhipicephalus spp., Rhizoglyphus spp.,
Sarcoptes spp., Tarsonemus spp. and Tetranychus spp.; and
of the order Thysanura, for example
Lepisma saccharina.

The good pesticidal activity of the compounds of formula I according to the invention corresponds to a mortality of at least 50-60 % of the mentioned pests.

The activity of the compounds of the invention and of the compositions comprising them can be substantially broadened and adapted to prevailing circumstances by the addition of other insecticides and/or acaricides. Examples of suitable additives include representatives of the following classes of compounds: organophosphorus compounds, nitrophenols and derivatives thereof, formamidines, ureas, carbamates, pyrethroids, chlorinated hydrocarbons, and Bacillus thuringiensis preparations.

The compounds of formula I are used in unmodified form or, preferably, together with the adjuvants conventionally employed in formulation technology, and can therefore be formulated in known manner e.g. into emulsifiable concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules, and also encapsulations in polymer substances. As with the compositions, the methods of application, such as spraying, atomising, dusting, scattering or pouring, are chosen in accordance with the intended objectives and the prevailing circumstances. The compounds of formula I are also suitable for use in the treatment of seed. For this purpose it is possible either to treat or dress the seed with the active ingredient or with a formulation comprising the active ingredient before sowing, or to apply the active ingredient into the seed furrow at the time of sowing.

The formulations, i.e. the compositions, preparations or mixtures comprising the

compound (active ingredient) of formula I, or combinations of those compounds with other insecticides or acaricides, and, where appropriate, a solid or liquid adjuvant, are prepared in known manner, e.g. by homogeneously mixing and/or grinding the active ingredients with extenders, e.g. solvents, solid carriers and, where appropriate, surface-active compounds (surfactants).

Suitable solvents are: aromatic hydrocarbons, preferably the C₈ to C₁₂ fractions of alkylbenzenes, e.g. xylene mixtures or alkylated naphthalenes, aliphatic or cycloaliphatic hydrocarbons such as cyclohexane, paraffins or tetrahydronaphthalene, alcohols such as ethanol, propanol or butanol, and glycols and their ethers and esters, such as propylene glycol, dipropylene glycol ether, ethylene glycol, ethylene glycol monomethyl or monoethyl ether, ketones such as cyclohexanone, isophorone or diacetone alcohol, strongly polar solvents such as N-methyl-2-pyrrolidone, dimethyl sulfoxide or dimethylformamide, or water, vegetable oils such as rape oil, castor oil, coconut oil or soybean oil; and, where appropriate, silicone oils.

The solid carriers used, e.g. for dusts and dispersible powders, are normally natural mineral fillers such as calcite, talcum, kaolin, montmorillonite or attapulgite. In order to improve the physical properties it is also possible to add highly dispersed silicic acids or highly dispersed absorbent polymers. Suitable granulated adsorptive carriers are porous types, for example pumice, broken brick, sepiolite or bentonite; and suitable nonsorbent carriers are, for example, calcite or sand. In addition, a great number of granulated materials of inorganic or organic nature can be used, e.g. especially dolomite or pulverised plant residues.

Depending on the nature of the compound of formula I to be formulated, or of the combinations of those compounds with other insecticides or acaricides, suitable surface-active compounds are non-ionic, cationic and/or anionic surfactants having good emulsifying, dispersing and wetting properties. The term "surfactants" will also be understood as comprising mixtures of surfactants.

Both so-called water-soluble soaps and water-soluble synthetic surface-active compounds are suitable anionic surfactants.

Suitable soaps are the alkali metal salts, alkaline earth metal salts or unsubstituted or substituted ammonium salts of higher fatty acids (C₁₀-C₂₂), e.g. the sodium or potassium

salts of oleic or stearic acid, or of natural fatty acid mixtures which can be obtained e.g. from coconut oil or tall oil. Mention may also be made of fatty acid methyltaurin salts.

More frequently, however, so-called synthetic surfactants are used, especially fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylarylsulfonates.

The fatty sulfonates or sulfates are usually in the form of alkali metal salts, alkaline earth metal salts or unsubstituted or substituted ammonium salts and generally contain a C₈-C₂₂alkyl radical, which also includes the alkyl moiety of acyl radicals, e.g. the sodium or calcium salt of lignosulfonic acid, of dodecyl sulfate or of a mixture of fatty alcohol sulfates obtained from natural fatty acids. These compounds also comprise the salts of sulfated and sulfonated fatty alcohol/ethylene oxide adducts. The sulfonated benzimidazole derivatives preferably contain 2 sulfonic acid groups and one fatty acid radical containing approximately 8 to 22 carbon atoms. Examples of alkylarylsulfonates are the sodium, calcium or triethanolamine salts of dodecylbenzenesulfonic acid, dibutynaphthalenesulfonic acid, or of a condensate of naphthalenesulfonic acid and formaldehyde. Also suitable are corresponding phosphates, e.g. salts of the phosphoric acid ester of an adduct of p-nonylphenol with 4 to 14 mol of ethylene oxide, or phospholipids.

Non-ionic surfactants are preferably polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, said derivatives containing 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon moiety and 6 to 18 carbon atoms in the alkyl moiety of the alkylphenols. Further suitable non-ionic surfactants are the water-soluble adducts of polyethylene oxide with polypropylene glycol, ethylenediaminopolypropylene glycol and alkylpolypropylene glycol containing 1 to 10 carbon atoms in the alkyl chain, which adducts contain 20 to 250 ethylene glycol ether groups and 10 to 100 propylene glycol ether groups. These compounds usually contain 1 to 5 ethylene glycol units per propylene glycol unit.

Representative examples of non-ionic surfactants are nonylphenolpolyethoxyethanols, castor oil polyglycol ethers, polypropylene/polyethylene oxide adducts, tributylphenoxy-polyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol. Fatty acid esters of polyoxyethylene sorbitan, e.g. polyoxyethylene sorbitan trioleate, are also suitable non-ionic surfactants.

Cationic surfactants are preferably quaternary ammonium salts which contain, as N-substituent, at least one C₈-C₂₂alkyl radical and, as further substituents, unsubstituted or halogenated lower alkyl, benzyl or hydroxy-lower alkyl radicals. The salts are preferably in the form of halides, methyl sulfates or ethyl sulfates, e.g. stearyltrimethylammonium chloride or benzylid(2-chloroethyl)ethylammonium bromide.

The surfactants customarily employed in formulation technology are described, for example, in the following publications:

"McCutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Glen Rock, NJ, USA, 1988",

H. Stache, "Tensid-Taschenbuch", 2nd edition, C. Hanser Verlag, Munich, Vienna, 1981,

M. and J. Ash, "Encyclopedia of Surfactants", Vol. I-III, Chemical Publishing Co., New York, 1980-1981.

The pesticidal compositions usually comprise 0.1 to 99 %, preferably 0.1 to 95 %, of a compound of formula I or combinations of that compound with other insecticides or acaricides, 1 to 99.9 % of a solid or liquid adjuvant, and 0 to 25 %, preferably 0.1 to 25 %, of a surfactant. Whereas commercial products will preferably be formulated as concentrates, the end user will normally employ dilute formulations comprising considerably lower active ingredient concentrations. Typical application concentrations are from 0.1 to 1000 ppm, preferably from 0.1 to 500 ppm. The rates of application per hectare are generally from 1 to 1000 g of active ingredient per hectare, preferably from 25 to 500 g/ha.

Preferred formulations have especially the following compositions (throughout, percentages are by weight), active ingredient being understood as meaning a compound of formula I:

Emulsifiable concentrates:

active ingredient:	1 to 90 %, preferably 5 to 20 %
surface-active agent:	1 to 30 %, preferably 10 to 20 %
liquid carrier:	5 to 94 %, preferably 70 to 85 %

Dusts:

active ingredient: 0.1 to 10 %, preferably 0.1 to 1 %
solid carrier: 99.9 to 90 %, preferably 99.9 to 99 %

Suspension concentrates:

active ingredient: 5 to 75 %, preferably 10 to 50 %
water: 94 to 24 %, preferably 88 to 30 %
surface-active agent: 1 to 40 %, preferably 2 to 30 %

Wettable powders:

active ingredient: 0.5 to 90 %, preferably 1 to 80 %
surface-active agent: 0.5 to 20 %, preferably 1 to 15 %
solid carrier: 5 to 95 %, preferably 15 to 90 %

Granules:

active ingredient: 0.5 to 30 %, preferably 3 to 15 %
solid carrier: 99.5 to 70 %, preferably 97 to 85 %

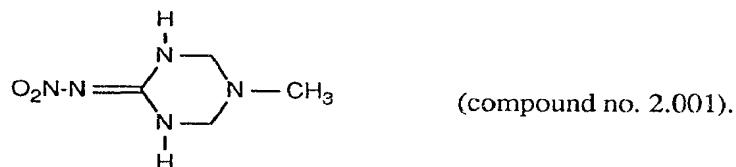
The compositions may also comprise further auxiliaries such as stabilisers, e.g. vegetable oils or epoxidised vegetable oils (epoxidised coconut oil, rape oil or soybean oil), anti-foams, e.g. silicone oil, preservatives, viscosity regulators, binders, tackifiers as well as fertilisers or other active ingredients for obtaining special effects.

The following Examples serve to illustrate the invention, but do not limit the invention.

Example 1 (Preparation of starting materials of formula IV):a) Preparation of 2-nitroimino-5-methyl-1,3,5-triazacyclohexane:

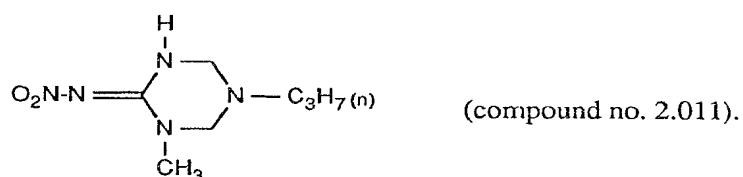
A mixture of 26.0 g of 2-nitroguanidine, 31.1 ml of an 8M solution of methylamine in ethanol, 38 ml of a 37 % solution of formaldehyde in water, and 100 ml of ethanol is heated at 50°C for 2 hours and then filtered. The crystals which have been filtered off are washed three times with 20 ml of ethanol each time and then dried, yielding the title compound, m.p. 173-175°C, of the formula

- 20 -



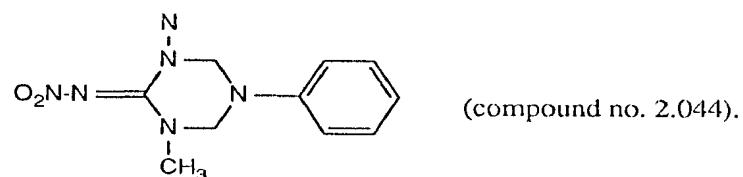
b) Preparation of 1-methyl-2-nitroimino-5-n-propyl-1,3,5-triazacyclohexane:

A mixture of 17.1 g of 1-methyl-2-nitroguanidine, 12.0 ml of n-propylamine, 22.0 ml of a 37 % solution of formaldehyde in water, and 40 ml of ethanol is heated at 50°C for 4 hours. A further 7.0 ml of n-propylamine and 13.0 ml of a 37 % solution of formaldehyde in water are then added. After stirring at 50°C for 2 hours, the reaction mixture is concentrated by evaporation in vacuo and the crystals which have separated out are stirred with ether, yielding 26.9 g of the title compound, m.p. 84-86°C, of the formula

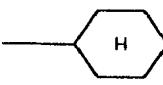
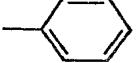
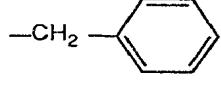
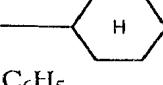
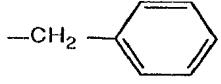


c) Preparation of 1-methyl-2-nitroimino-5-phenyl-1,3,5-triazacyclohexane:

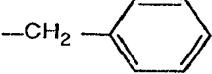
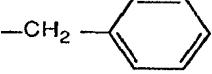
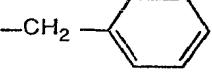
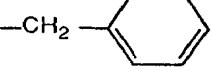
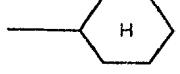
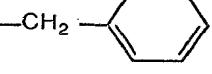
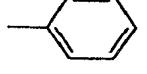
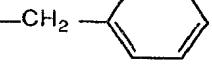
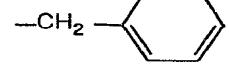
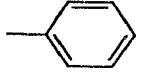
Three drops of concentrated hydrochloric acid are added to a mixture of 2.36 g of 1-methyl-2-nitroguanidine, 2.11 ml of aniline and 1.80 g of paraformaldehyde in 30 ml of toluene, and the mixture is then boiled in a water separator for 6 hours. The reaction mixture is then concentrated by evaporation in vacuo and the resulting crude product is recrystallised from methanol, yielding the title compound, m.p. 169-172°C, of the formula

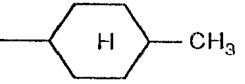
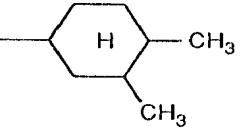
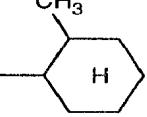


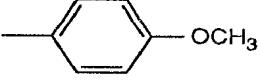
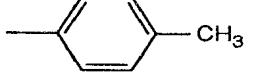
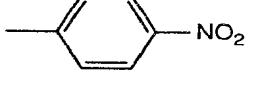
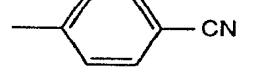
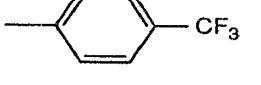
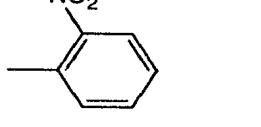
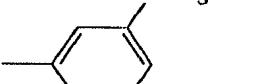
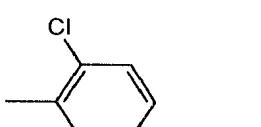
The following compounds of formula IV can be prepared as indicated above:

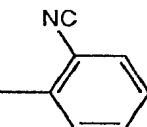
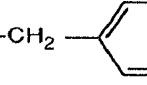
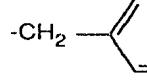
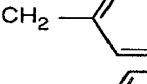
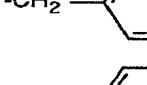
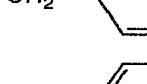
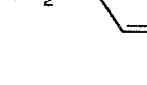
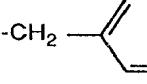
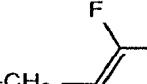
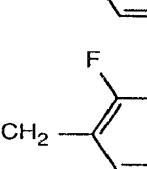
Comp. No.	R ₂	R ₃	Phys. data
2.001	H	CH ₃	m.p. 173-175°C
2.002	H	-C ₂ H ₅	m.p. 181-182°C
2.003	H	-C ₃ H ₇ (n)	
2.004	H	CH(CH ₃) ₂	
2.005	H		m.p. 225-227°C
2.006	H		
2.007	H		
2.008	H		
2.009	CH ₃	-CH ₃	m.p. 134-135°C
2.010	CH ₃	-C ₂ H ₅	m.p. 112°C
2.011	CH ₃	-C ₃ H ₇ (n)	m.p. 84-86°C
2.012	CH ₃	-CH ₂ (CH ₃) ₂	m.p. 154°C
2.013	CH ₃		m.p. 177°C
2.014	CH ₃		m.p. 103-104°C
2.015	CH ₃	-C ₆ H ₅	m.p. 169-172°C
2.016	CH ₃		m.p. 161-163°C
2.017	-C ₂ H ₅	-CH ₃	
2.018	-C ₂ H ₅	-C ₂ H ₅	m.p. 95-96°C
2.019	-C ₂ H ₅	-C ₃ H ₇ (n)	
2.020	-C ₂ H ₅	-CH(CH ₃) ₂	

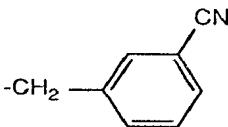
Comp. No.	R ₂	R ₃	Phys. data
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2.022	-C ₂ H ₅		
2.023	-C ₂ H ₅		
2.024	-C ₂ H ₅		
2.025		-CH ₃	
2.026		-C ₂ H ₅	m.p. 138-139°C
2.027		-C ₃ H ₇ (n)	
2.028		-CH(CH ₃) ₂	
2.029			
2.030			
2.031			
2.032			
2.033		-CH ₃	m.p. 109-111°C
2.034		-C ₂ H ₅	

Comp. No.	R ₂	R ₃	Phys. data
2.035	-CH ₂ - 	-C ₃ H ₇ (n)	
2.036	-CH ₂ - 	CH(CH ₃) ₂	
2.037	-CH ₂ - 		
2.038	-CH ₂ - 		
2.039	-CH ₂ - 		
2.040	-CH ₂ - 	-CH ₂ - 	
2.041	H	-CH ₂ -COOCH ₃	
2.042	-CH ₃	-CH ₂ -COOCH ₃	
2.043		-CH ₂ -COOCH ₃	
2.044	-CH ₃		m.p. 169-172°C
2.045	-CH ₃	-CH ₂ CF ₃	
2.046	-CH ₃	-CH ₂ CH ₂ F	
2.047	-CH ₃	-CH ₂ CH ₂ Br	
2.048	-CH ₃	-CH ₂ CH ₂ CH ₂ Cl	
2.049	-CH ₃	-CH ₂ CH ₂ CH ₂ Br	
2.050	-CH ₃	-CH ₂ CH ₂ Cl	
2.051	-CH ₃	-CH ₂ CH(Cl)CH ₂ CH ₂ CH ₂ Cl	
2.052	-CH ₃	-CH ₂ CH ₂ OH	m.p. 121-123°C
2.053	-CH ₃	-CH ₂ CH ₂ CH ₂ OH	
2.054	-CH ₃	-CH ₂ CH ₂ CH ₂ CH ₂ OH	m.p. 81-83°C

Comp. No.	R ₂	R ₃	Phys. data
2.055	-CH ₃	-CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ OH	
2.056	-CH ₃	-CH(CH ₃)CH ₃ OH	
2.057	-CH ₃	-CH(C ₂ H ₅)CH ₂ OH	
2.058	-CH ₃	-CH ₂ CH(CH ₃)OH	
2.059	-CH ₃	-CH ₂ CH(OH)CH ₂ OH	
2.060	-CH ₃	-CH(CH ₂ OH) ₂	
2.061	-CH ₃	-CH ₂ CH ₂ OCH ₃	
2.062	-CH ₃	-CH ₂ CH ₂ CH ₂ OC ₂ H ₅	
2.063	-CH ₃	-CH(CH ₃)CH ₂ OCH ₃	
2.064	-CH ₃	-CH ₂ CH(OCH ₃) ₂	
2.065	-CH ₃	-CH ₂ CH(OC ₂ H ₅) ₂	
2.066	-CH ₃	-CH ₂ CH ₂ N(CH ₃) ₂	
2.067	-CH ₃	-CH ₂ CH ₂ N(C ₂ H ₅) ₂	
2.068	-CH ₃	-CH ₂ CH ₂ CH ₂ N(CH ₃) ₂	
2.069	-CH ₃	-CH ₂ CH ₂ CH ₂ N(C ₂ H ₅) ₂	
2.070	-CH ₃	-CH ₂ COOC ₂ H ₅	
2.071	-CH ₃	CH ₂ CH ₂ COOC ₂ H ₅	m.p. 110-112°C
2.072	-CH ₃	-CH(CH ₃)CH ₂ COOC ₂ H ₅	
2.073	-CH ₃	-CH(CH ₂ OH)COOCH ₃	
2.074	-CH ₃		
2.075	-CH ₃		m.p. 151-153°C (cis isomer)
			m.p. 138-140°C (trans isomer)
2.076	-CH ₃		
2.077	-CH ₃		

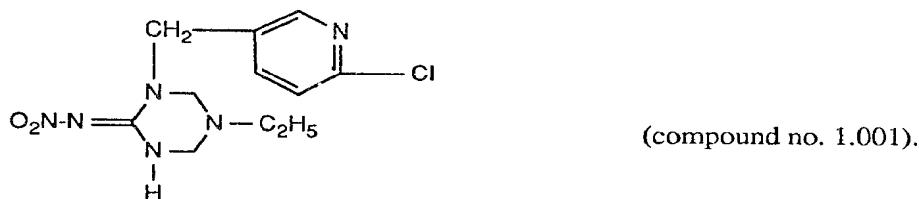
Comp. No.	R ₂	R ₃	Phys. data
2.078	-CH ₃	-CH ₂ -CH=CH ₂	m.p. 53-55°C
2.079	-CH ₃		
2.080	-CH ₃		m.p. 170-173°C
2.081	-CH ₃		m.p. 174-176°C
2.082	-CH ₃		m.p. 195-197°C
2.083	-CH ₃		m.p. 230°C
2.084	-CH ₃		m.p. 222-226°C
2.085	-CH ₃		m.p. 163-166°C
2.086	-CH ₃		
2.087	-CH ₃		
2.088	-CH ₃		

Comp. No.	R ₂	R ₃	Phys. data
2.089	-CH ₃		
2.090	-CH ₃	-CH ₂ - 	m.p. 235-238°C
2.091	-CH ₃	-CH ₂ - 	m.p. 143-145°C
2.092	-CH ₃	-CH ₂ - 	m.p. 132-134°C
2.093	-CH ₃	-CH ₂ - 	m.p. 160-162°C
2.094	-CH ₃	-CH ₂ - 	m.p. 161-163°C
2.095	-CH ₃	-CH ₂ - 	m.p. 160-162°C
2.096	-CH ₃	-CH ₂ - 	
2.097	-CH ₃	-CH ₂ - 	
2.098	-CH ₃	-CH ₂ - 	

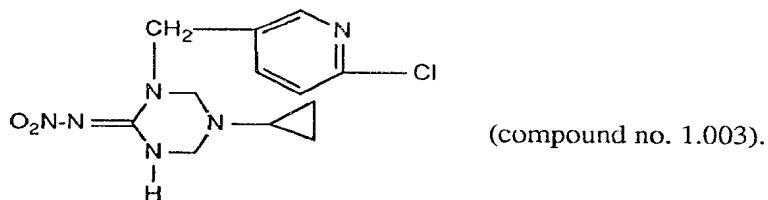
Comp. No.	R ₂	R ₃	Phys. data
2.099	-CH ₃	-CH ₂ - 	

Example 2:a) Preparation of 1-(2-chloropyrid-5-ylmethyl)-2-nitroimino-5-ethyl-1,3,5-triazacyclohexane:

A mixture of 1.15 g of 1-(2-chloropyrid-5-ylmethyl)-2-nitroguanidine, 0.75 ml of a 37 % solution of formaldehyde in water, 0.32 ml of a 70 % solution of ethylamine in water, and 5 ml of ethanol is heated at 50°C for 4 hours. The reaction mixture is then concentrated by evaporation in vacuo, the residue is suspended in 20 ml of ethanol, and the resulting crystals are filtered off, yielding the title compound, m.p. 125-126°C, of the formula

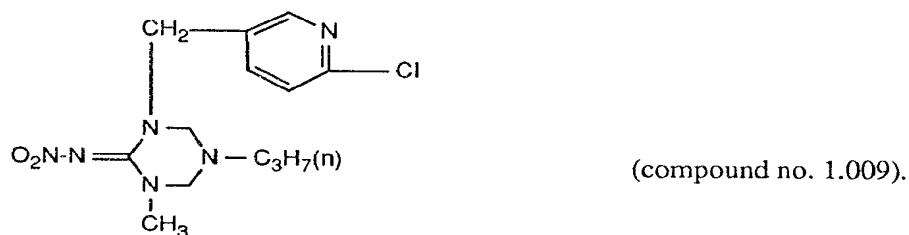
b) Preparation of 1-(2-chloropyrid-5-ylmethyl)-2-nitroimino-5-cyclopropyl-1,3,5-triazacyclohexane:

A mixture of 2.96 g of 2-nitroimino-5-cyclopropyl-1,3,5-triazacyclohexane, 2.59 g of 2-chloro-5-chloromethylpyridine and 2.43 g of potassium carbonate in 60 ml of acetonitrile is heated under reflux for 16 hours. The resulting reaction mixture is filtered, the filtrate is concentrated by evaporation in vacuo and the residue that forms is chromatographed on silica gel with dichloromethane/ethyl acetate (1:1), yielding the title compound, m.p. 125-127°C, of the formula



c) Preparation of 1-(2-chloropyrid-5-ylmethyl)-2-nitroimino-3-methyl-5-n-propyl-1,3,5-triazacyclohexane:

A mixture of 20.1 g of 1-methyl-2-nitroimino-5-n-propyl-1,3,5-triazacyclohexane, 16.2 g of 2-chloro-5-chloromethylpyridine, 0.17 g of caesium chloride and 27.7 g of potassium carbonate in 150 ml of DMF*) is heated at 110°C for 9 hours and then filtered over Celite. The filtrate is concentrated by evaporation in vacuo. The resulting crude product is dissolved in 200 ml of dichloromethane and washed with 100 ml of water and 100 ml of saturated sodium chloride solution, dried over magnesium sulfate and then concentrated by evaporation. The residue is recrystallised from ethyl acetate, yielding the title compound, m.p. 137-138°C, of the formula



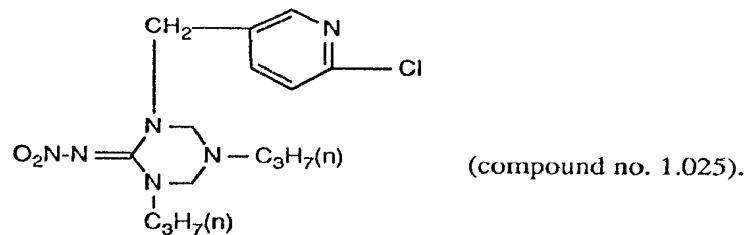
*)dimethylformamide

d) Preparation of 1-(2-chloropyrid-5-ylmethyl)-2-nitroimino-3,5-di-(n-propyl)-1,3,5-triazacyclohexane:

0.30 g of sodium hydride (80 % in white oil) is added to a solution of 3.12 g of 1-(2-chloropyrid-5-ylmethyl)-2-nitroimino-5-n-propyl-1,3,5-triazacyclohexane in 50 ml of acetonitrile. After stirring the reaction mixture at room temperature for 3 hours, 1.8 ml of n-propyl iodide are added, and the reaction mixture is then stirred at room temperature for 16 hours and at 80°C for 2 hours. The residue obtained after concentration by evaporation in vacuo is taken up in 100 ml of ethyl acetate, washed with 50 ml of saturated sodium chloride solution, dried over magnesium sulfate and again concentrated by evaporation.

- 29 -

The crystals obtained as residue are recrystallised at 0°C from ethyl acetate, yielding the title compound, m.p. 112-113°C, of the formula



The following compounds of formula I can be prepared as indicated above:

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.001		H	H	-C ₂ H ₅	m.p. 125-126°C
1.002		H	H	-C ₃ H ₇ (n)	m.p. 115-117°C
1.003		H	H		m.p. 125-127°C
1.004		H	H		m.p. 150-151°C
1.005		H	H		m.p. 143-145°C
1.006		H	H		m.p. 108-110°C
1.007		H	CH ₃	-CH ₃	amorphous mass
1.008		H	CH ₃	-C ₂ H ₅	m.p. 124-125°C
1.009		H	CH ₃	-C ₃ H ₇ (n)	m.p. 137-138°C
1.010		H	CH ₃	-CH(CH ₃) ₂	
1.011		H	-CH ₃		m.p. 104-106°C
1.012		H	-CH ₃		m.p. 146-147°C

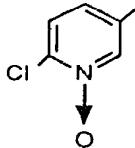
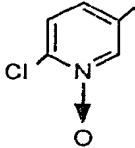
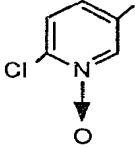
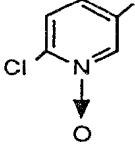
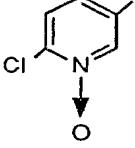
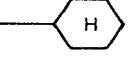
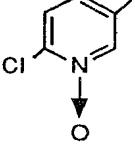
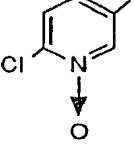
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.013		H	-CH ₃		m.p. 146-149°C
1.014		H	-CH ₃		m.p. 116-118°C
1.015		H	-C ₂ H ₅		
1.016		H	-C ₂ H ₅		m.p. 113-114°C
1.017		H	-C ₂ H ₅		
1.018		H	-C ₂ H ₅		
1.019		H	-C ₂ H ₅		
1.020		H	-C ₂ H ₅		
1.021		H	-C ₂ H ₅		
1.022		H	-C ₂ H ₅		
1.023		H	-C ₃ H ₇ (n)		
1.024		H	-C ₃ H ₇ (n)		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.025		H	-C ₃ H ₇ (n)	-C ₃ H ₇ (n)	m.p. 112-113°C
1.026		H	-C ₃ H ₇ (n)	-CH(CH ₃) ₂	
1.027		H	-C ₃ H ₇ (n)		
1.028		H	-C ₃ H ₇ (n)		
1.029		H	-C ₃ H ₇ (n)		
1.030		H	-C ₃ H ₇ (n)	-CH ₂	
1.031		H	-CH(CH ₃) ₂	-CH ₃	
1.032		H	-CH(CH ₃) ₂	-C ₂ H ₅	
1.033		H	-CH(CH ₃) ₂	-C ₃ H ₇ (n)	
1.034		H	-CH(CH ₃) ₂	-CH(CH ₃) ₂	
1.035		H	-CH(CH ₃) ₂		
1.036		H	-CH(CH ₃) ₂		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.037		H	-CH(CH ₃) ₂		
1.038		H	-CH(CH ₃) ₂		
1.039		H		CH ₃	
1.040		H		-C ₂ H ₅	m.p. 115-116°C
1.041		H		-C ₃ H ₇ (n)	
1.042		H		-CH(CH ₃) ₂	
1.043		H			
1.044		H			
1.045		H			
1.046		H			
1.047		H	H		-CH ₃

- 34 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.048		H	H	-C ₂ H ₅	
1.049		H	H	-C ₃ H ₇ (n)	
1.050		H	H	-CH(CH ₃) ₂	
1.051		H	H		
1.052		H	H		
1.053		H	H		
1.054		H	H	-CH ₂ - 	

- 35 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.055		H	CH ₃	-CH ₃	
1.056		H	CH ₃	-C ₂ H ₅	m.p. 152-155°C
1.057		H	CH ₃	-C ₃ H ₇ (n)	m.p. 117-121°C
1.058		H	CH ₃	-CH(CH ₃) ₂	m.p. 138°C
1.059		H	CH ₃		
1.060		H	CH ₃		m.p. 153°C
1.061		H	CH ₃		

- 36 -

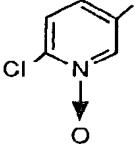
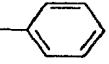
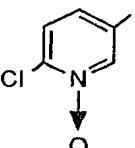
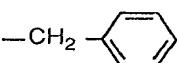
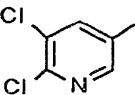
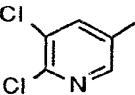
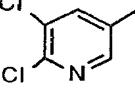
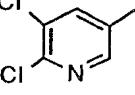
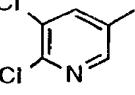
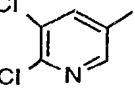
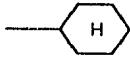
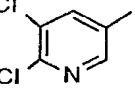
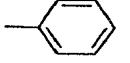
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.062		H	CH ₃		-CH ₂ -
1.063		H			-CH ₃
1.064		H			-C ₂ H ₅
1.065		H			-C ₃ H _{7(n)}
1.066		H			-CH(CH ₃) ₂
1.067		H			
1.068		H			

- 37 -

Comp. A
No.R₁ R₂R₃

Phys. data

1.069		H		
1.070		H		
1.071		H H		-CH ₃
1.072		H H		-C ₂ H ₅
1.073		H H		-C ₃ H _{7(n)}
1.074		H H		-CH(CH ₃) ₂
1.075		H H		
1.076		H H		
1.077		H H		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.078		H	H		
1.079		H	CH ₃		-CH ₃
1.080		H	CH ₃		-C ₂ H ₅
1.081		H	CH ₃		-C ₃ H ₇ (n)
1.082		H	CH ₃		-CH(CH ₃) ₂
1.083		H	CH ₃		
1.084		H	CH ₃		
1.085		H	CH ₃		
1.086		H	CH ₃		
1.087		H			-CH ₃

- 39 -

Comp. No.	A	R_1	R_2	R_3	Phys. data
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1.088		H			-C ₂ H ₅
1.089		H			-C ₃ H ₇ (n)
1.090		H			-CH(CH ₃) ₂
1.091		H			
1.092		H			
1.093		H			
1.094		H			-CH ₂ -
1.095		H	H		-C ₂ H ₅
1.096		H	H		-C ₃ H ₇ (n)
1.097		H	H		-CH(CH ₃) ₂
1.098		H	H		
1.099		H	H		

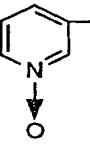
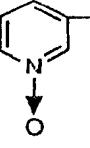
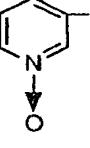
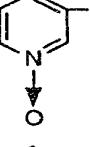
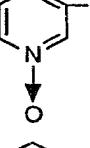
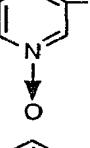
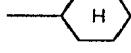
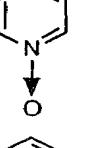
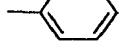
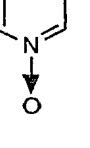
- 40 -

Comp. A No.	R_1	R_2	R_3	Phys. data
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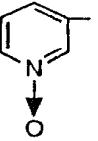
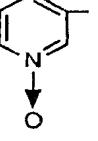
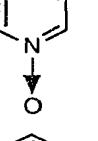
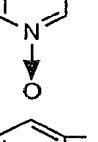
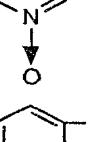
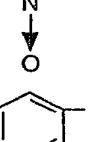
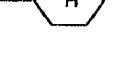
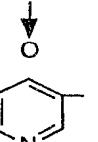
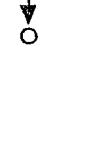
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1.101		H H		
1.102		H CH ₃		
1.103		H CH ₃		
1.104		H CH ₃		amorphous
1.105		H CH ₃		
1.106		H CH ₃		
1.107		H CH ₃		
1.108		H CH ₃		
1.109		H CH ₃		
1.110		H		
1.111		H		
1.112		H		
1.113		H		
1.114		H		
1.115		H		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.116		H			
1.117		H			
1.118		H	H		-CH ₃
1.119		H	H		-C ₂ H ₅
1.120		H	H		-C ₃ H ₇ (n)
1.121		H	H		-CH(CH ₃) ₂
1.122		H	H		
1.123		H	H		
1.124		H	H		
1.125		H	H		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.126		H	H	-CH ₃	
1.127		H	CH ₃	-C ₂ H ₅	
1.128		H	CH ₃	-C ₃ H ₇ (n)	amorphous mass
1.129		H	CH ₃	-CH(CH ₃) ₂	
1.130		H	CH ₃		m.p. 185°C
1.131		H	CH ₃		
1.132		H	CH ₃		
1.133		H	CH ₃	 -CH ₂ - 	

Comp. No.	A	R ₁ R ₂	R ₃	Phys. data
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1.134	 	H — 	-CH ₃	
1.135	 	H — 	-C ₂ H ₅	
1.136	 	H — 	-C ₃ H ₇ (n)	
1.137	 	H — 	-CH(CH ₃) ₂	
1.138	 	H — 		
1.139	 	H — 		
1.140	 	H — 		
1.141	 	H — 	-CH ₂ — 	

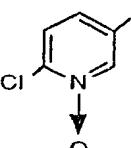
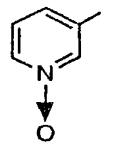
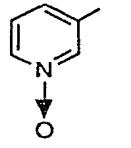
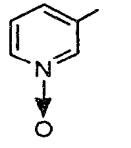
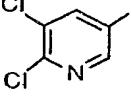
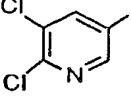
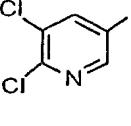
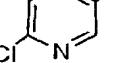
- 44 -

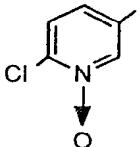
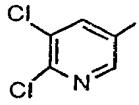
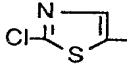
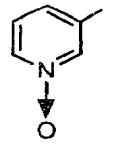
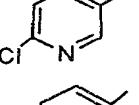
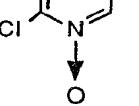
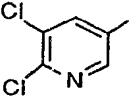
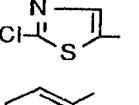
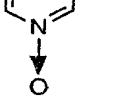
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.142		H	H	-CH ₃	m.p. 142-144°C
1.143		H	-CH ₃	-CH ₃	
1.144		H		-CH ₃	
1.145		H	H	-C ₂ H ₅	
1.146		H	-CH ₃	-C ₂ H ₅	
1.147		H		-C ₂ H ₅	
1.148		H	-CH ₂ -	-C ₃ H ₇ (n)	m.p. 127-129°C
1.149		H	-CH ₂ -	-C ₃ H ₇ (n)	
1.150		H	-CH ₂ -	-C ₃ H ₇ (n)	
1.151		H	-CH ₂ -	-C ₃ H ₇ (n)	
1.152		H	-CH ₂ -		

- 45 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.153		H	-CH ₂ -		m.p. 190-192°C
1.154		H	-CH ₂ -		
1.155		H	H	-CH ₂ COOCH ₃	m.p. 184-186°C
1.156		H	CH ₃	-CH ₂ COOCH ₃	m.p. 185°C
1.157		H		-CH ₂ COOCH ₃	
1.158		H	H	-CH ₂ COOCH ₃	
1.159		H	CH ₃	-CH ₂ COOCH ₃	
1.160		H		-CH ₂ COOCH ₃	
1.161		H	H	-CH ₂ COOCH ₃	
1.162		H	CH ₃	-CH ₂ COOCH ₃	

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.163		H			-CH ₂ COOCH ₃
1.164		H	H		-CH ₂ COOCH ₃
1.165		H	CH ₃		-CH ₂ COOCH ₃
1.166		H			-CH ₂ COOCH ₃
1.167		H	H		-CH ₂ COOCH ₃
1.168		H	CH ₃		-CH ₂ COOCH ₃
1.169		H			-CH ₂ COOCH ₃
1.170		H	CH ₃		-CH ₂ CF ₃

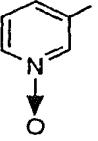
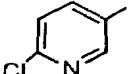
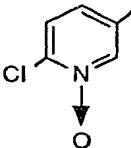
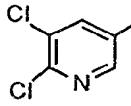
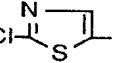
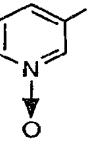
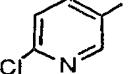
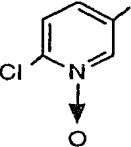
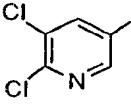
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1.171		H	CH ₃	-CH ₂ CF ₃	
1.172		H	CH ₃	-CH ₂ CF ₃	
1.173		H	CH ₃	-CH ₂ CF ₃	
1.174		H	CH ₃	-CH ₂ CF ₃	
1.175		H	CH ₃	-CH ₂ CH ₂ F	
1.176		H	CH ₃	-CH ₂ CH ₂ F	
1.177		H	CH ₃	-CH ₂ CH ₂ F	
1.178		H	CH ₃	-CH ₂ CH ₂ F	
1.179		H	CH ₃	-CH ₂ CH ₂ F	

- 48 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.180		H	CH ₃		-CH ₂ CH ₂ Br
1.181		H	CH ₃		-CH ₂ CH ₂ Br
1.182		H	CH ₃		-CH ₂ CH ₂ Br
1.183		H	CH ₃		-CH ₂ CH ₂ Br
1.184		H	CH ₃		-CH ₂ CH ₂ Br
1.185		H	CH ₃		-CH ₂ CH ₂ CH ₂ Cl
1.186		H	CH ₃		-CH ₂ CH ₂ CH ₂ Cl
1.187		H	CH ₃		-CH ₂ CH ₂ CH ₂ Cl
1.188		H	CH ₃		-CH ₂ CH ₂ CH ₂ Cl

- 49 -

Comp. No.	A	R_1	R_2	R_3	Phys. data
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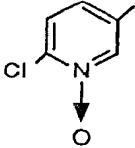
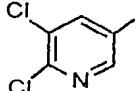
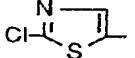
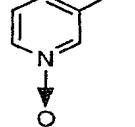
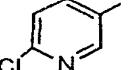
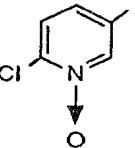
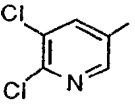
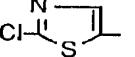
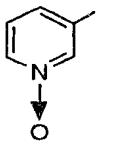
1.189		H	CH_3	- $CH_2CH_2CH_2Cl$	
1.190		H	CH_3	- $CH_2CH_2CH_2Br$	
1.191		H	CH_3	- $CH_2CH_2CH_2Br$	
1.192		H	CH_3	- $CH_2CH_2CH_2Br$	
1.193		H	CH_3	- $CH_2CH_2CH_2Br$	
1.194		H	CH_3	- $CH_2CH_2CH_2Br$	
1.195		H	CH_3	- CH_2CH_2Cl	
1.196		H	CH_3	- CH_2CH_2Cl	
1.197		H	CH_3	- CH_2CH_2Cl	

- 50 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.198		H	CH ₃	-CH ₂ CH ₂ Cl	
1.199		H	CH ₃	-CH ₂ CH ₂ Cl	
1.200		H	CH ₃	-CH ₂ CH(Cl)CH ₂ CH ₂ CH ₂ Cl	
1.201		H	CH ₃	-CH ₂ CH(Cl)CH ₂ CH ₂ CH ₂ Cl	
1.202		H	CH ₃	-CH ₂ CH(Cl)CH ₂ CH ₂ CH ₂ Cl	
1.203		H	CH ₃	-CH ₂ CH(Cl)CH ₂ CH ₂ CH ₂ Cl	
1.204		H	CH ₃	-CH ₂ CH(Cl)CH ₂ CH ₂ CH ₂ Cl	
1.205		H	CH ₃	-CH ₂ CH ₂ OH	amorphous mass
1.206		H	CH ₃	-CH ₂ CH ₂ OH	

- 51 -

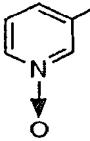
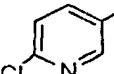
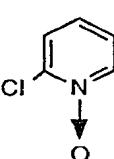
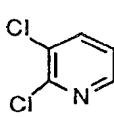
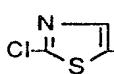
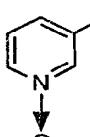
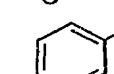
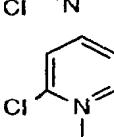
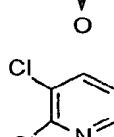
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.207		H	CH ₃		-CH ₂ CH ₂ OH
1.208		H	CH ₃		-CH ₂ CH ₂ OH
1.209		H	CH ₃		-CH ₂ CH ₂ OH
1.210		H	CH ₃	-CH ₂ CH ₂ CH ₂ OH	amorphous mass
1.211		H	CH ₃	-CH ₂ CH ₂ CH ₂ OH	
1.212		H	CH ₃		-CH ₂ CH ₂ CH ₂ OH
1.213		H	CH ₃		-CH ₂ CH ₂ CH ₂ OH
1.214		H	CH ₃		-CH ₂ CH ₂ CH ₂ OH
1.215		H	CH ₃	-CH ₂ (CH ₂) ₂ CH ₂ OH	m.p. 108-110°C

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.216		H	CH ₃		-CH ₂ (CH ₂) ₂ CH ₂ OH
1.217		H	CH ₃		-CH ₂ (CH ₂) ₂ CH ₂ OH
1.218		H	CH ₃		-CH ₂ (CH ₂) ₂ CH ₂ OH
1.219		H	CH ₃		-CH ₂ (CH ₂) ₂ CH ₂ OH
1.220		H	CH ₃		-CH ₂ (CH ₂) ₃ CH ₂ OH
1.221		H	CH ₃		-CH ₂ (CH ₂) ₃ CH ₂ OH
1.222		H	CH ₃		-CH ₂ (CH ₂) ₃ CH ₂ OH
1.223		H	CH ₃		-CH ₂ (CH ₂) ₃ CH ₂ OH
1.224		H	CH ₃		-CH ₂ (CH ₂) ₃ CH ₂ OH

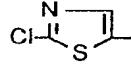
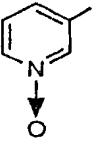
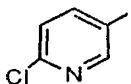
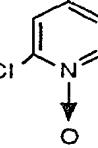
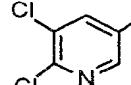
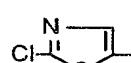
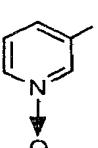
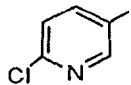
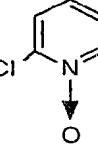
- 53 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.225		H	CH ₃	-CH(CH ₃)CH ₂ OH	amorphous mass
1.226		H	CH ₃	-CH(CH ₃)CH ₂ OH	
1.227		H	CH ₃	-CH(CH ₃)CH ₂ OH	
1.228		H	CH ₃	-CH(CH ₃)CH ₂ OH	
1.229		H	CH ₃	-CH(CH ₃)CH ₂ OH	
1.230		H	CH ₃	-CH(C ₂ H ₅)CH ₂ OH	
1.231		H	CH ₃	-CH(C ₂ H ₅)CH ₂ OH	
1.232		H	CH ₃	-CH(C ₂ H ₅)CH ₂ OH	
1.233		H	CH ₃	-CH(C ₂ H ₅)CH ₂ OH	

Comp. No.	A	R_1	R_2	R_3	Phys. data
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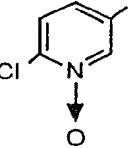
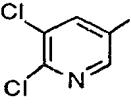
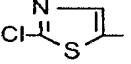
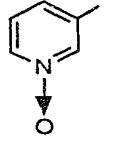
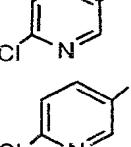
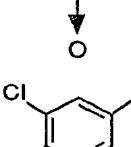
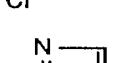
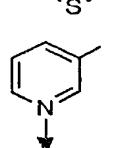
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1.235		H	CH ₃	-	CH ₂ CH(CH ₃)OH
1.236		H	CH ₃	-	CH ₂ CH(CH ₃)OH
1.237		H	CH ₃	-	CH ₂ CH(CH ₃)OH
1.238		H	CH ₃	-	CH ₂ CH(CH ₃)OH
1.239		H	CH ₃	-	CH ₂ CH(CH ₃)OH
1.240		H	CH ₃	-	CH ₂ CH(OH)CH ₂ OH
1.241		H	CH ₃	-	CH ₂ CH(OH)CH ₂ OH
1.242		H	CH ₃	-	CH ₂ CH(OH)CH ₂ OH

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.243		H	CH ₃	-CH ₂ CH(OH)CH ₂ OH	
1.244		H	CH ₃	-CH ₂ CH(OH)CH ₂ OH	
1.245		H	CH ₃	-CH(CH ₂ OH) ₂	
1.246		H	CH ₃	-CH(CH ₂ OH) ₂	
1.247		H	CH ₃	-CH(CH ₂ OH) ₂	
1.248		H	CH ₃	-CH(CH ₂ OH) ₂	
1.249		H	CH ₃	-CH(CH ₂ OH) ₂	
1.250		H	CH ₃	-CH ₂ CH ₂ OCH ₃	
1.251		H	CH ₃	-CH ₂ CH ₂ OCH ₃	

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.252		H	CH ₃		-CH ₂ CH ₂ OCH ₃
1.253		H	CH ₃		-CH ₂ CH ₂ OCH ₃
1.254		H	CH ₃		-CH ₂ CH ₂ OCH ₃
1.255		H	CH ₃		-CH ₂ CH ₂ CH ₂ OC ₂ H ₅
1.256		H	CH ₃		-CH ₂ CH ₂ CH ₂ OC ₂ H ₅
1.257		H	CH ₃		-CH ₂ CH ₂ CH ₂ OC ₂ H ₅
1.258		H	CH ₃		-CH ₂ CH ₂ CH ₂ OC ₂ H ₅
1.259		H	CH ₃		-CH ₂ CH ₂ CH ₂ OC ₂ H ₅
1.260		H	CH ₃		-CH(CH ₃)CH ₂ OCH ₃

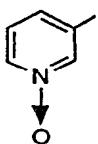
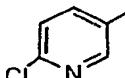
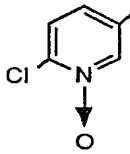
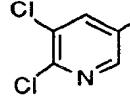
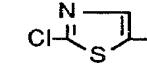
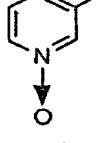
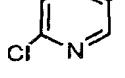
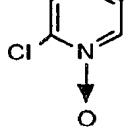
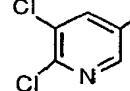
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.261		H	CH ₃		-CH(CH ₃)CH ₂ OCH ₃
1.262		H	CH ₃		-CH(CH ₃)CH ₂ OCH ₃
1.263		H	CH ₃		-CH(CH ₃)CH ₂ OCH ₃
1.264		H	CH ₃		-CH(CH ₃)CH ₂ OCH ₃
1.265		H	CH ₃		-CH ₂ CH(OCH ₃) ₂
1.266		H	CH ₃		-CH ₂ CH(OCH ₃) ₂
1.267		H	CH ₃		-CH ₂ CH(OCH ₃) ₂
1.268		H	CH ₃		-CH ₂ CH(OCH ₃) ₂
1.269		H	CH ₃		-CH ₂ CH(OCH ₃) ₂

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.270		H	CH ₃		-CH ₂ CH(OC ₂ H ₅) ₂
1.271		H	CH ₃		-CH ₂ CH(OC ₂ H ₅) ₂
1.272		H	CH ₃		-CH ₂ CH(OC ₂ H ₅) ₂
1.273		H	CH ₃		-CH ₂ CH(OC ₂ H ₅) ₂
1.274		H	CH ₃		-CH ₂ CH(OC ₂ H ₅) ₂
1.275		H	CH ₃		-CH ₂ CH ₂ N(CH ₃) ₂
1.276		H	CH ₃		-CH ₂ CH ₂ N(CH ₃) ₂
1.277		H	CH ₃		-CH ₂ CH ₂ N(CH ₃) ₂
1.278		H	CH ₃		-CH ₂ CH ₂ N(CH ₃) ₂

Comp. No.	A	R ₁ R ₂	R ₃	Phys. data
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1.279		H CH ₃	-CH ₂ CH ₂ N(CH ₃) ₂
1.280		H CH ₃	-CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.281		H CH ₃	-CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.282		H CH ₃	-CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.283		H CH ₃	-CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.284		H CH ₃	-CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.285		H CH ₃	-CH ₂ CH ₂ CH ₂ N(CH ₃) ₂
1.286		H CH ₃	-CH ₂ CH ₂ CH ₂ N(CH ₃) ₂
1.287		H CH ₃	-CH ₂ CH ₂ CH ₂ N(CH ₃) ₂

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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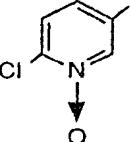
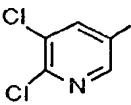
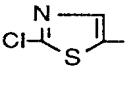
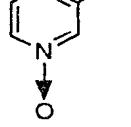
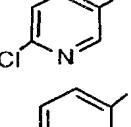
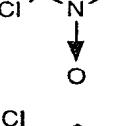
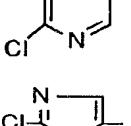
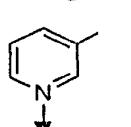
1.288		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(CH ₃) ₂
1.289		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(CH ₃) ₂
1.290		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.291		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.292		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.293		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.294		H	CH ₃	-	-CH ₂ CH ₂ CH ₂ N(C ₂ H ₅) ₂
1.295		H	CH ₃	-CH ₂ COOC ₂ H ₅	amorphous mass
1.296		H	CH ₃	-CH ₂ COOC ₂ H ₅	

Comp. No.	A	R ₁ R ₂	R ₃	Phys. data
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1.297		H CH ₃	-CH ₂ COOC ₂ H ₅	
1.298		H CH ₃	-CH ₂ COOC ₂ H ₅	
1.299		H CH ₃	-CH ₂ COOC ₂ H ₅	
1.300		H CH ₃	-CH ₂ CH ₂ COOC ₂ H ₅	m.p. 78-80°C
1.301		H CH ₃	-CH ₂ CH ₂ COOC ₂ H ₅	
1.302		H CH ₃	-CH ₂ CH ₂ COOC ₂ H ₅	
1.303		H CH ₃	-CH ₂ CH ₂ COOC ₂ H ₅	
1.304		H CH ₃	-CH ₂ CH ₂ COOC ₂ H ₅	
1.305		H CH ₃	-CH(CH ₃)CH ₂ COOC ₂ H ₅	

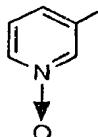
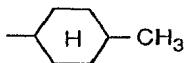
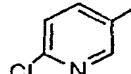
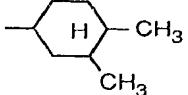
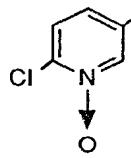
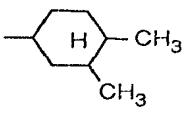
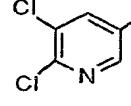
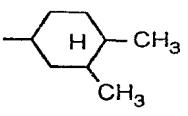
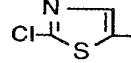
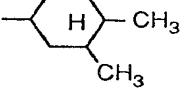
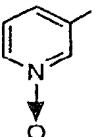
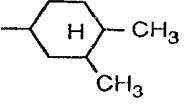
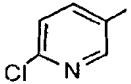
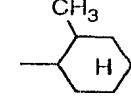
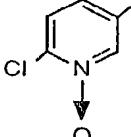
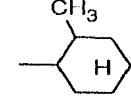
- 62 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.306		H	CH ₃		-CH(CH ₃)CH ₂ COOC ₂ H ₅
1.307		H	CH ₃		-CH(CH ₃)CH ₂ COOC ₂ H ₅
1.308		H	CH ₃		-CH(CH ₃)CH ₂ COOC ₂ H ₅
1.309		H	CH ₃		-CH(CH ₃)CH ₂ COOC ₂ H ₅
1.310		H	CH ₃		-CH(CH ₂ OH)COOCH ₃
1.311		H	CH ₃		-CH(CH ₂ OH)COOCH ₃
1.312		H	CH ₃		-CH(CH ₂ OH)COOCH ₃
1.313		H	CH ₃		-CH(CH ₂ OH)COOCH ₃
1.314		H	CH ₃		-CH(CH ₂ OH)COOCH ₃

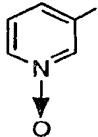
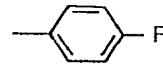
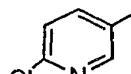
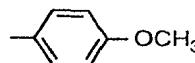
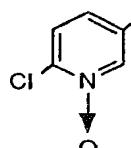
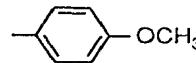
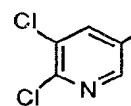
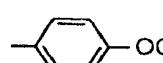
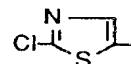
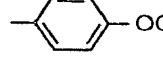
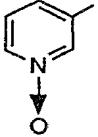
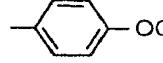
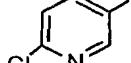
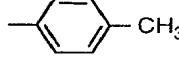
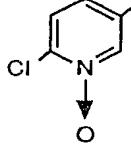
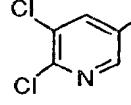
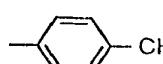
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.315		H	CH ₃		
1.316		H	CH ₃		
1.317		H	CH ₃		
1.318		H	CH ₃		
1.319		H	CH ₃		
1.320		H	CH ₃		m.p. 137-139°C (cis isomer) m.p. 170-172°C (trans isomer)
1.321		H	CH ₃		
1.322		H	CH ₃		
1.323		H	CH ₃		

Comp. A	R_1	R_2	R_3	Phys. data
No.				

1.324		H	CH ₃	
1.325		H	CH ₃	
1.326		H	CH ₃	
1.327		H	CH ₃	
1.328		H	CH ₃	
1.329		H	CH ₃	
1.330		H	CH ₃	
1.331		H	CH ₃	

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.332		H	CH ₃		
1.333		H	CH ₃		
1.334		H	CH ₃		
1.335		H	CH ₃	-CH ₂ CH=CH ₂	m.p. 75-77°C
1.336		H	CH ₃	-CH ₂ CH=CH ₂	
1.337		H	CH ₃	-CH ₂ CH=CH ₂	
1.338		H	CH ₃	-CH ₂ CH=CH ₂	
1.339		H	CH ₃	-CH ₂ CH=CH ₂	
1.340		H	CH ₃		

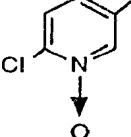
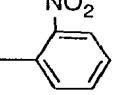
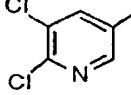
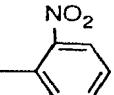
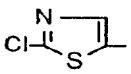
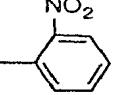
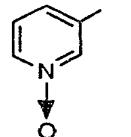
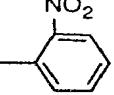
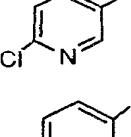
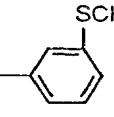
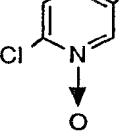
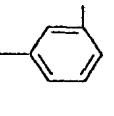
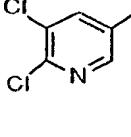
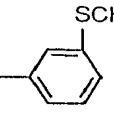
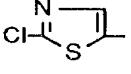
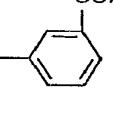
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.341		H	CH ₃		
1.342		H	CH ₃		
1.343		H	CH ₃		
1.344		H	CH ₃		
1.345		H	CH ₃		amorphous mass
1.346		H	CH ₃		
1.347		H	CH ₃		
1.348		H	CH ₃		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.349		H	CH ₃		
1.350		H	CH ₃		m.p. 204°C
1.351		H	CH ₃		
1.352		H	CH ₃		
1.353		H	CH ₃		
1.354		H	CH ₃		
1.355		H	CH ₃		amorphous mass
1.356		H	CH ₃		
1.357		H	CH ₃		

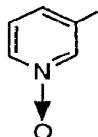
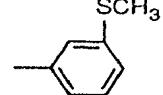
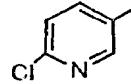
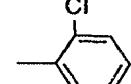
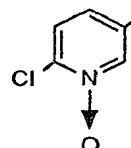
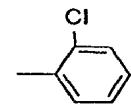
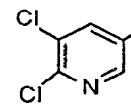
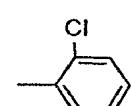
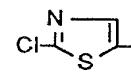
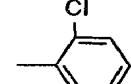
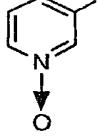
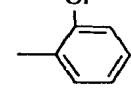
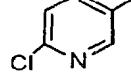
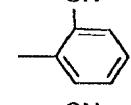
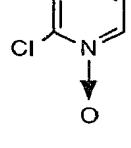
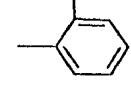
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.358		H	CH ₃		
1.359		H	CH ₃		
1.360		H	CH ₃		m.p. 219°C
1.361		H	CH ₃		
1.362		H	CH ₃		
1.363		H	CH ₃		
1.364		H	CH ₃		
1.365		H	CH ₃		amorphous mass
1.366		H	CH ₃		

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.367		H	CH ₃		
1.368		H	CH ₃		
1.369		H	CH ₃		
1.370		H	CH ₃		amorphous mass
1.371		H	CH ₃		
1.372		H	CH ₃		
1.373		H	CH ₃		
1.374		H	CH ₃		
1.375		H	CH ₃		

Comp. No.	A	R_1	R_2	R_3	Phys. data
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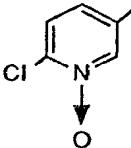
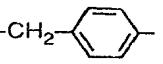
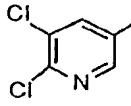
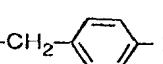
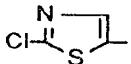
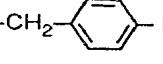
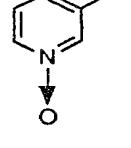
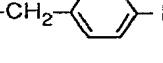
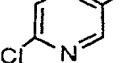
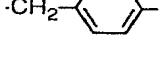
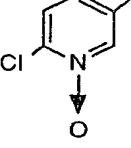
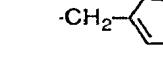
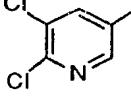
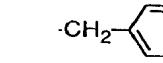
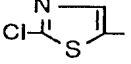
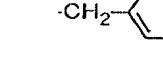
1.376		H	CH_3		
1.377		H	CH_3		
1.378		H	CH_3		
1.379		H	CH_3		
1.380		H	CH_3		
1.381		H	CH_3		
1.382		H	CH_3		
1.383		H	CH_3		

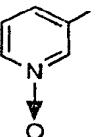
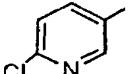
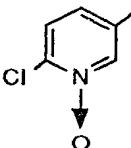
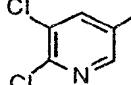
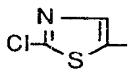
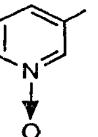
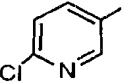
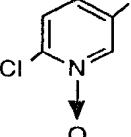
Comp. No.	A	R_1	R_2	R_3	Phys. data
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1.384		H	CH_3		
1.385		H	CH_3		
1.386		H	CH_3		
1.387		H	CH_3		
1.388		H	CH_3		
1.389		H	CH_3		
1.390		H	CH_3		
1.391		H	CH_3		

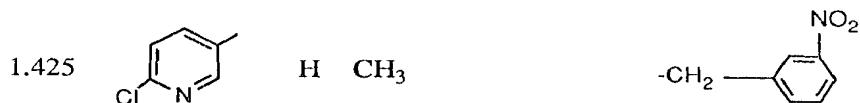
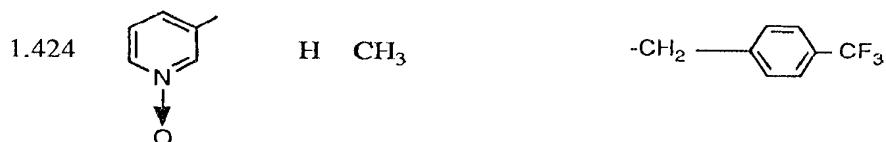
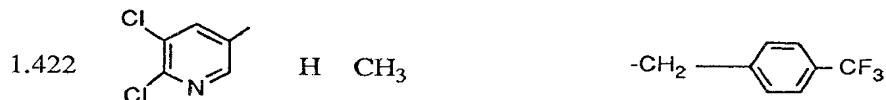
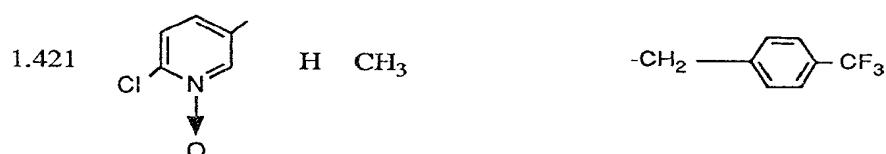
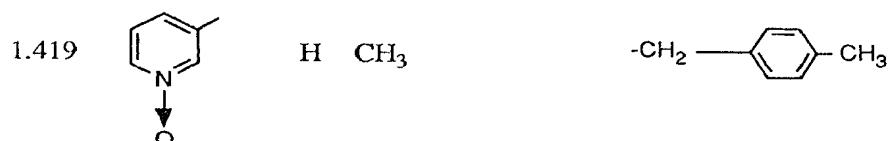
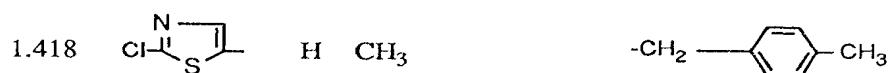
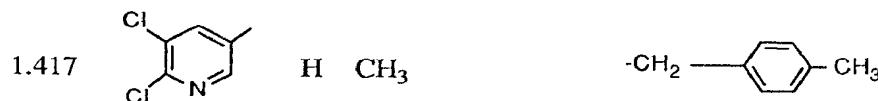
Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.392		H	CH ₃		
1.393		H	CH ₃		
1.394		H	CH ₃		
1.395		H	CH ₃	-CH ₂ -	amorphous mass
1.396		H	CH ₃	-CH ₂ -	
1.397		H	CH ₃	-CH ₂ -	
1.398		H	CH ₃	-CH ₂ -	
1.399		H	CH ₃	-CH ₂ -	
1.400		H	CH ₃	-CH ₂ -	m.p. 162-164°C

Comp. No.	A	R ₁ R ₂	R ₃	Phys. data
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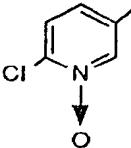
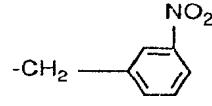
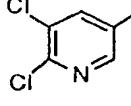
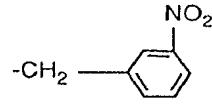
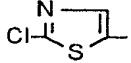
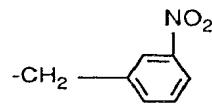
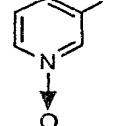
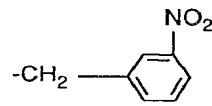
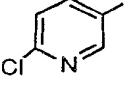
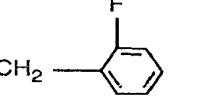
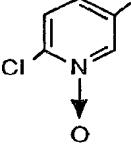
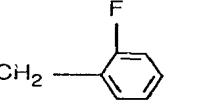
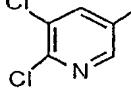
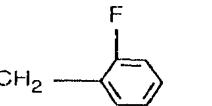
1.401		H CH ₃	-CH ₂ - 	
1.402		H CH ₃	-CH ₂ - 	
1.403		H CH ₃	-CH ₂ - 	
1.404		H CH ₃	-CH ₂ - 	
1.405		H CH ₃	-CH ₂ - 	m.p. 125-127°C
1.406		H CH ₃	-CH ₂ - 	
1.407		H CH ₃	-CH ₂ - 	
1.408		H CH ₃	-CH ₂ - 	

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.409		H	CH ₃	-CH ₂ -C ₆ H ₄ -OCH ₃	
1.410		H	CH ₃	-CH ₂ -C ₆ H ₄ -Cl	m.p. 147-149°C
1.411		H	CH ₃	-CH ₂ -C ₆ H ₄ -Cl	
1.412		H	CH ₃	-CH ₂ -C ₆ H ₄ -Cl	
1.413		H	CH ₃	-CH ₂ -C ₆ H ₄ -Cl	
1.414		H	CH ₃	-CH ₂ -C ₆ H ₄ -Cl	
1.415		H	CH ₃	-CH ₂ -C ₆ H ₄ -CH ₃	m.p. 155-157°C
1.416		H	CH ₃	-CH ₂ -C ₆ H ₄ -CH ₃	

Comp. A No.	R_1	R_2	R_3	Phys. data
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Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
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1.426		H	CH ₃		
1.427		H	CH ₃		
1.428		H	CH ₃		
1.429		H	CH ₃		
1.430		H	CH ₃		
1.431		H	CH ₃		
1.432		H	CH ₃		

Comp. No.	A	R_1	R_2	R_3	Phys. data
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1.433		H	CH_3		
1.434		H	CH_3		
1.435		H	CH_3		
1.436		H	CH_3		
1.437		H	CH_3		
1.438		H	CH_3		
1.439		H	CH_3		

- 78 -

Comp. No.	A	R ₁	R ₂	R ₃	Phys. data
1.440		H	CH ₃		
1.441		H	CH ₃		
1.442		H	CH ₃		
1.443		H	CH ₃		
1.444		H	CH ₃		
1.445		H	CH ₃	-C ₃ H ₇ (n)	m.p. 157-158°C

- 79 -

Example 3:Formulations (throughout, percentages are by weight)

<u>Example F1: Emulsifiable concentrates</u>	a)	b)	c)
a compound of Example 2	25 %	40 %	50 %
calcium dodecylbenzenesulfonate	5 %	8 %	6 %
castor oil polyethylene glycol ether (36 mol of ethylene oxide)	5 %	-	-
tributylphenol polyethylene glycol ether (30 mol of ethylene oxide)	-	12 %	4 %
cyclohexanone	-	15 %	20 %
xylene mixture	65 %	25 %	20 %

Emulsions of any desired concentration can be produced from such concentrates by dilution with water.

<u>Example F2: Solutions</u>	a)	b)	c)	d)
a compound of Example 2	80 %	10 %	5 %	95 %
ethylene glycol monomethyl ether	20 %	-	-	-
polyethylene glycol (mol. wt. 400)	-	70 %	-	-
N-methyl-2-pyrrolidone	-	20 %	-	-
epoxidised coconut oil petroleum fraction (boiling range 160-190°C)	-	-	1 %	5 %
	-	-	94 %	-

The solutions are suitable for application in the form of micro-drops.

<u>Example F3: Granules</u>	a)	b)	c)	d)
a compound of Example 2	5 %	10 %	8 %	21 %
kaolin	94 %	-	79 %	54 %
highly dispersed silicic acid	1 %	-	13 %	7 %
attapulgite	-	90 %	-	18 %

The active ingredient is dissolved in methylene chloride, the solution is sprayed onto the carrier, and the solvent is subsequently evaporated off in vacuo.

Example F4: Dusts

	a)	b)
a compound of Example 2	2 %	5 %
highly dispersed silicic acid	1 %	5 %
talcum	97 %	-
kaolin	-	90 %

Ready-for-use dusts are obtained by intimately mixing the carriers with the active ingredient.

Example F5: Wettable powders

	a)	b)	c)
a compound of Example 2	25 %	50 %	75 %
sodium lignosulfonate	5 %	5 %	-
sodium laurylsulfate	3 %	-	5 %
sodium diisobutylnaphthalene-sulfonate	-	6 %	10 %
octylphenol polyethylene glycol ether (7-8 mol of ethylene oxide)	-	2 %	-
highly dispersed silicic acid	5 %	10 %	10 %
kaolin	62 %	27 %	-

The active ingredient or active ingredient combination is mixed with the adjuvants and the mixture is thoroughly ground in a suitable mill, affording wettable powders which can be diluted with water to give suspensions of any desired concentration.

Example F6: Emulsifiable concentrate

a compound of Example 2	10 %
octylphenol polyethylene glycol ether (4-5 mol of ethylene oxide)	3 %
calcium dodecylbenzenesulfonate	3 %
castor oil polyglycol ether (36 mol of ethylene oxide)	4 %
cyclohexanone	30 %

xylene mixture	50 %
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Emulsions of any desired concentration can be obtained from this concentrate by dilution with water.

Example F7: Dusts

	a)	b)
a compound of Example 2	5 %	8 %
talcum	95 %	-
kaolin	-	92 %

Ready-for-use dusts are obtained by mixing the active ingredient with the carrier and grinding the mixture in a suitable mill.

Example F8: Extruder granules

a compound of Example 2	10 %
sodium lignosulfonate	2 %
carboxymethylcellulose	1 %
kaolin	87 %

The active ingredient or active ingredient combination is mixed and ground with the adjuvants, and the mixture is moistened with water. The mixture is extruded, granulated and then dried in a stream of air.

Example F9: Coated granules

a compound of Example 2	3 %
polyethylene glycol (mol. wt. 200)	3 %
kaolin	94 %

The finely ground active ingredient or active ingredient combination is uniformly applied, in a mixer, to the kaolin moistened with polyethylene glycol. Non-dusty coated granules are obtained in this manner.

Example F10: Suspension concentrate

a compound of Example 2	40 %
ethylene glycol	10 %
nonylphenol polyethylene glycol	

- 82 -

ether (15 mol of ethylene oxide)	6 %
sodium lignosulfonate	10 %
carboxymethylcellulose	1 %
silicone oil in the form of a 75 % aqueous emulsion	1 %
water	32 %

The finely ground active ingredient or active ingredient combination is intimately mixed with the adjuvants, giving a suspension concentrate from which suspensions of any desired concentration can be obtained by dilution with water.

Example 4: Action against Nilaparvata lugens

Rice plants are sprayed with an aqueous emulsion comprising 400 ppm of test compound. After the spray coating has dried, the rice plants are populated with cicada larvae in the 2nd and 3rd stages. Evaluation is made 21 days later. The percentage reduction in the population (% activity) is determined by comparing the number of surviving cicadas on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against Nilaparvata lugens in this test. In particular, compounds 1.007, 1.008, 1.009, 1.011, 1.012, 1.013, 1.014, 1.056, 1.057, 1.058, 1.104, 1.128, 1.130, 1.156, 1.205, 1.215, 1.300, 1.320, 1.335 and 1.410 are more than 80-90 % effective.

Example 5: Action against Nephrotettix cincticeps

Rice plants are sprayed with an aqueous emulsion comprising 400 ppm of test compound. After the spray coating has dried, the rice plants are populated with cicada larvae in the 2nd and 3rd stages. Evaluation is made 21 days later. The percentage reduction in the population (% activity) is determined by comparing the number of surviving cicadas on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against Nephrotettix cincticeps in this test. In particular, compounds 1.007, 1.008, 1.009, 1.011, 1.012, 1.013, 1.014, 1.056, 1.057, 1.058, 1.060, 1.104, 1.128, 1.156, 1.205, 1.215, 1.300, 1.320, 1.335 and 1.410 are more than 80 % effective.

Example 6: Action against *Myzus persicae*

Pea seedlings are infested with *Myzus persicae* and then sprayed with a spray mixture comprising 400 ppm of the test compound, and incubated at 20°C. Evaluation is made 3 and 6 days later. The percentage reduction in the population (% activity) is determined by comparing the number of dead aphids on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against *Myzus persicae* in this test. In particular, compounds 1.001, 1.002, 1.006, 1.007, 1.008, 1.009, 1.011, 1.012, 1.104 and 1.156 are more than 80 % effective.

Example 7: Action against *Aphis craccivora*

Pea seedlings are infested with *Aphis craccivora* and then sprayed with a spray mixture comprising 400 ppm of the test compound, and incubated at 20°C. Evaluation is made 3 and 6 days later. The percentage reduction in the population (% activity) is determined by comparing the number of dead aphids on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against *Aphis craccivora* in this test. In particular, compounds 1.001, 1.002, 1.006, 1.007, 1.008, 1.009, 1.011, 1.012, 1.104, 1.155, 1.156, 1.205 and 1.300 are more than 80 % effective.

Example 8: Systemic action against *Nilaparvata lugens*

Pots containing rice plants are placed in an aqueous emulsion solution comprising 400 ppm of the test compound. The rice plants are then populated with larvae in the 2nd and 3rd stages. Evaluation is made 6 days later. The percentage reduction in the population (% activity) is determined by comparing the number of cicadas on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against *Nilaparvata lugens* in this test. In particular, compounds 1.001, 1.002, 1.003, 1.004, 1.006, 1.007, 1.008, 1.009, 1.011, 1.012, 1.013, 1.014, 1.057, 1.058, 1.060, 1.104, 1.128, 1.130, 1.142, 1.156, 1.205, 1.215, 1.300, 1.320, 1.335, 1.410 and 1.445 are more than 80 % effective.

Example 9: Systemic action against *Nephrotettix cincticeps*

Pots containing rice plants are placed in an aqueous emulsion solution comprising 400 ppm of the test compound. The rice plants are then populated with larvae in the 2nd and 3rd stages. Evaluation is made 6 days later. The percentage reduction in the

population (% activity) is determined by comparing the number of cicadas on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against *Nephrotettix cincticeps* in this test. In particular, compounds 1.001, 1.002, 1.003, 1.004, 1.005, 1.006, 1.008, 1.009, 1.011, 1.012, 1.013, 1.014, 1.057, 1.058, 1.060, 1.104, 1.156 and 1.335 are more than 80 % effective.

Example 10: Systemic action against *Myzus persicae*

Pea seedlings are infested with *Myzus persicae* and then placed with their roots in a spray mixture comprising 400 ppm of the test compound, and incubated at 20°C. Evaluation is made 3 and 6 days later. The percentage reduction in the population (% activity) is determined by comparing the number of dead aphids on the treated plants with that on untreated plants.

Compounds of Example 2 exhibit good activity against *Myzus persicae* in this test. In particular, compounds 1.001, 1.002, 1.007, 1.008, 1.009, 1.011, 1.012, 1.025, 1.104 and 1.156 are more than 80 % effective.

Example 11: Action against *Bemisia tabaci*

Dwarf bean plants are placed in gauze cages and populated with adults of *Bemisia tabaci* (whitefly). When oviposition has taken place, all the adults are removed and 10 days later the plants and the nymphs located thereon are sprayed with an aqueous emulsion of the test compounds (concentration 400 ppm). Evaluation is made 14 days after application of the test compound by determining the % hatching rate in comparison with untreated controls.

Compounds of Example 2 exhibit good activity against *Bemisia tabaci* in this test. In particular, compounds 1.001, 1.002, 1.006, 1.007, 1.008, 1.009, 1.011, 1.012, 1.013, 1.014, 1.016, 1.104, 1.156 and 1.335 are more than 80 % effective.

Example 12: Action against *Ctenocephalus felis*

20 to 25 cat flea eggs (*Ctenocephalus felis*) are placed in each of a number of horizontal 50 ml cell culture bottles into which 15 g of a flea larvae nutrient medium comprising 100 ppm of the test compound have been introduced beforehand. The bottles are sealed and placed in an incubator at 26-27°C and 60-70 % humidity. After an incubation period

of 21 days, the development of adult fleas, unhatched pupae and larvae is assessed.

Compounds of Example 2 exhibit good activity in this test. In particular, compounds 1.009 and 1.025 are more than 80 % effective.

Example 13: Action against Blattella germanica

An amount of a 0.1 % solution of the test compound in acetone sufficient to produce a concentration of 1 g/m² is introduced into a petri dish having a diameter of 10 cm. When the solvent has evaporated, 10 Blattella germanica nymphs (final nymph stage) are placed in the dish so prepared and subjected to the action of the test compound for 2 hours. The nymphs are then narcotised with carbon dioxide, placed in a fresh petri dish and kept in the dark at 25°C and about 70 % humidity. The insecticidal action is evaluated 48 hours later by determining the mortality rate.

Compounds of Example 2 exhibit good activity in the above test. In particular, compound 1.104 is more than 60 % effective.

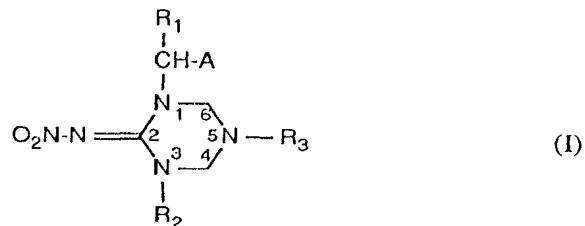
Example 14: Action against Boophilus microplus

Adult female ticks which are replete with blood are affixed to a PVC plate and covered with a cotton wool swab. For treatment, 10 ml of an aqueous test solution comprising 125 ppm of the test compound are poured over the test insects. The cotton wool swab is then removed and the ticks are incubated for 4 weeks until oviposition has taken place. The action against Boophilus microplus manifests itself either as mortality or sterility of the females or as ovicidal action in the eggs.

Compounds of Example 2 exhibit good activity against Boophilus microplus. In particular, compounds 1.008, 1.009 and 1.025 are more than 60 % effective in this test.

What is claimed is:

1. A compound of formula I



wherein

R₁ is hydrogen or C₁-C₄alkyl;

R₂ is hydrogen, C₁-C₆alkyl, C₃-C₆cycloalkyl or a radical -CH₂B;

R₃ is hydrogen; C₁-C₁₀alkyl; C₃-C₆cycloalkyl; C₁-C₁₀alkyl substituted by from 1 to 12

radicals from the group halogen, hydroxy, C₁-C₄alkoxy, C₁-C₄haloalkoxy having from 1 to 9 halogen atoms, di-(C₁-C₄alkyl)amino and C₁-C₅alkoxycarbonyl;

C₃-C₆cycloalkyl substituted by from 1 to 4 C₁-C₄alkyl radicals or halogen atoms;

C₂-C₈alkenyl or C₂-C₈alkynyl; C₂-C₈alkenyl or C₂-C₈alkynyl each of which is substituted by from 1 to 6 halogen atoms; phenyl; benzyl; or phenyl or benzyl each of which is substituted by from 1 to 3 ring substituents from the group halogen,

C₁-C₄alkyl, C₁-C₄haloalkyl having from 1 to 9 halogen atoms, C₁-C₄alkoxy,

C₁-C₄haloalkoxy having from 1 to 9 halogen atoms, C₁-C₄alkylthio, nitro and cyano;

- A is an unsubstituted or mono- to tetra-substituted aromatic or non-aromatic, monocyclic or bicyclic heterocyclic radical that can have one or two substituents from the group C₁-C₃haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl having from 1 to 3 halogen atoms, C₂-C₃alkenyl, C₂-C₃alkynyl, C₂-C₃haloalkenyl and C₂-C₃haloalkynyl each having from 1 to 4 halogen atoms, C₁-C₃haloalkoxy having from 1 to 7 halogen atoms, C₁-C₃alkylthio, C₁-C₃haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, and from one to four substituents from the group C₁-C₃alkyl, C₁-C₃alkoxy and halogen; and
- B is phenyl; cyanophenyl; nitrophenyl; halophenyl having from 1 to 3 halogen atoms;

phenyl substituted by C_1 - C_3 alkyl, C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, C_1 - C_3 alkoxy or by C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms; 3-pyridyl; 5-thiazolyl; 5-thiazoli substituted by one or two substituents from the group C_1 - C_3 alkyl, C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, C_2 - C_3 alkenyl, C_2 - C_3 alkynyl, C_1 - C_3 alkoxy, C_2 - C_3 haloalkenyl and C_2 - C_3 haloalkynyl each having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, halogen, cyano and nitro; or 3-pyridyl substituted by one or two radicals from the group C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, C_2 - C_3 alkenyl, C_2 - C_3 alkynyl, C_2 - C_3 haloalkenyl and C_2 - C_3 haloalkynyl each having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, or by from one to four radicals from the group C_1 - C_3 alkyl, C_1 - C_3 alkoxy and halogen;

or a salt thereof with an inorganic acid.

2. A compound of formula I according to claim 1 wherein R_3 is C_5 - C_{10} alkyl; C_3 - C_6 cycloalkyl; C_1 - C_{10} alkyl substituted by from 1 to 12 radicals from the group halogen, hydroxy, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy having from 1 to 9 halogen atoms, di-(C_1 - C_4 -alkyl)amino and C_1 - C_5 alkoxycarbonyl; C_3 - C_6 cycloalkyl substituted by from 1 to 4 C_1 - C_4 -alkyl radicals or halogen atoms; C_2 - C_8 alkenyl or C_2 - C_8 alkynyl; C_2 - C_8 alkenyl or C_2 - C_8 alkynyl each of which is substituted by from 1 to 6 halogen atoms; phenyl; benzyl; or phenyl or benzyl each of which is substituted by from 1 to 3 ring substituents from the group halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl having from 1 to 9 halogen atoms, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy having from 1 to 9 halogen atoms, C_1 - C_4 alkylthio, nitro and cyano; and R_1 , R_2 and A are as defined in claim 1, or a salt thereof with an inorganic acid.

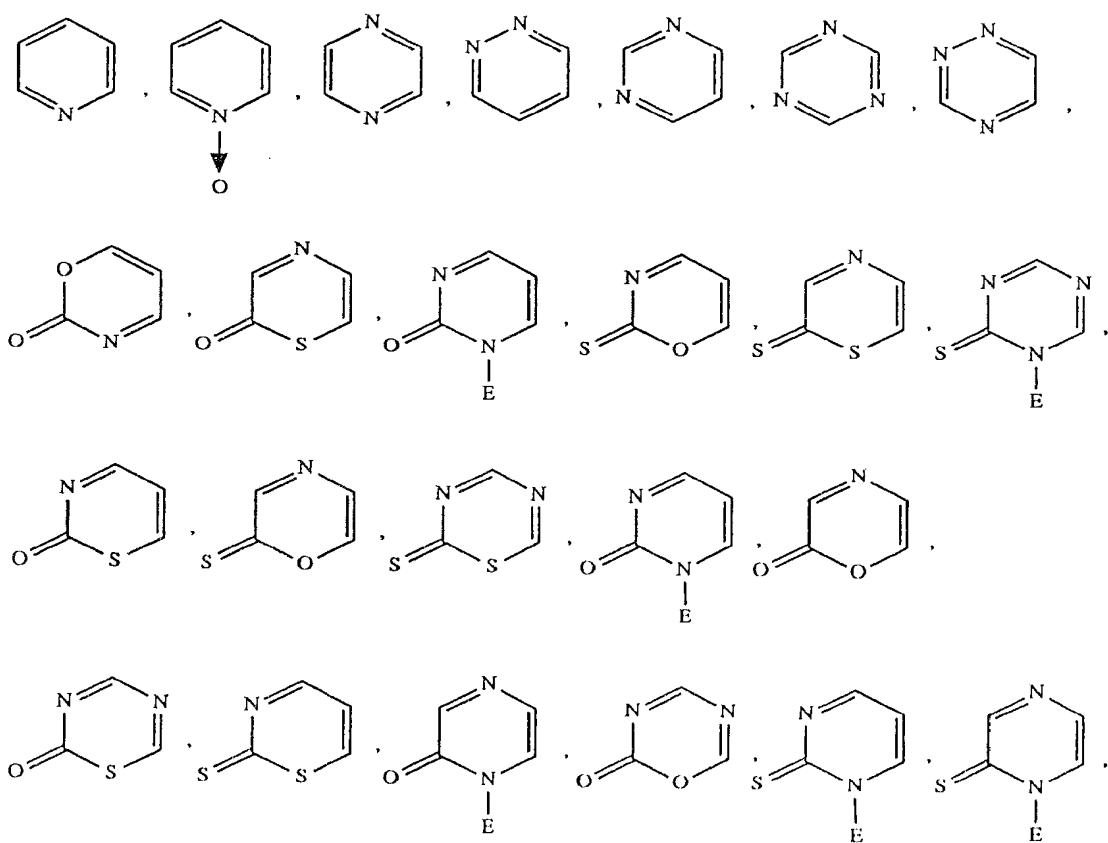
3. A compound according to either claim 1 or claim 2 wherein the heterocyclic radical A is unsaturated, is bonded via a carbon atom to the radical of the molecule of the compound of formula I and contains at least one nitrogen atom.

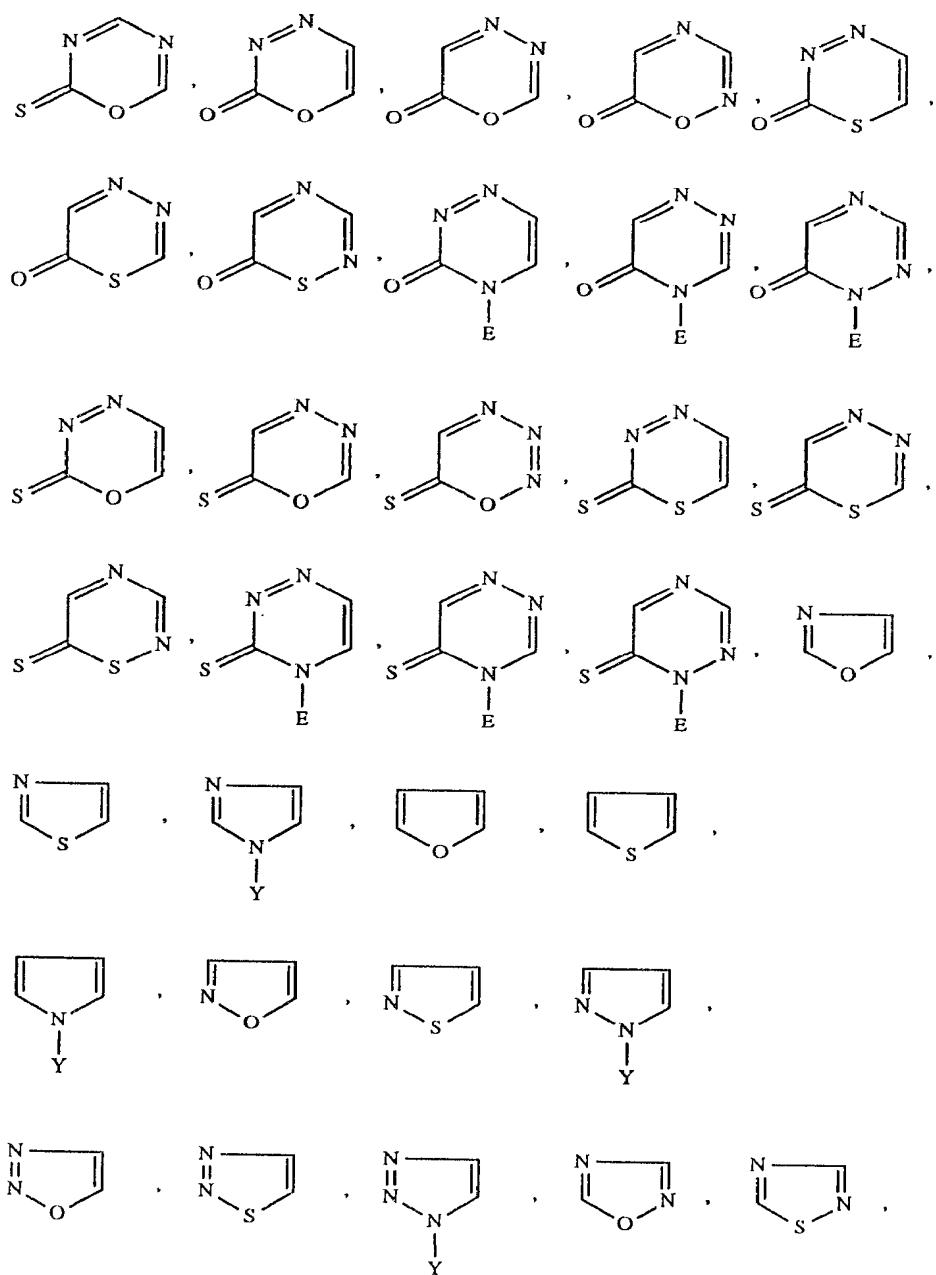
4. A compound according to claim 3 wherein the heterocyclic radical A is unsaturated, is

bonded via a carbon atom to the radical of the molecule of the compound of formula I and contains from one to three hetero atoms from the group oxygen, sulfur and nitrogen, not more than one oxygen or sulfur atom being present.

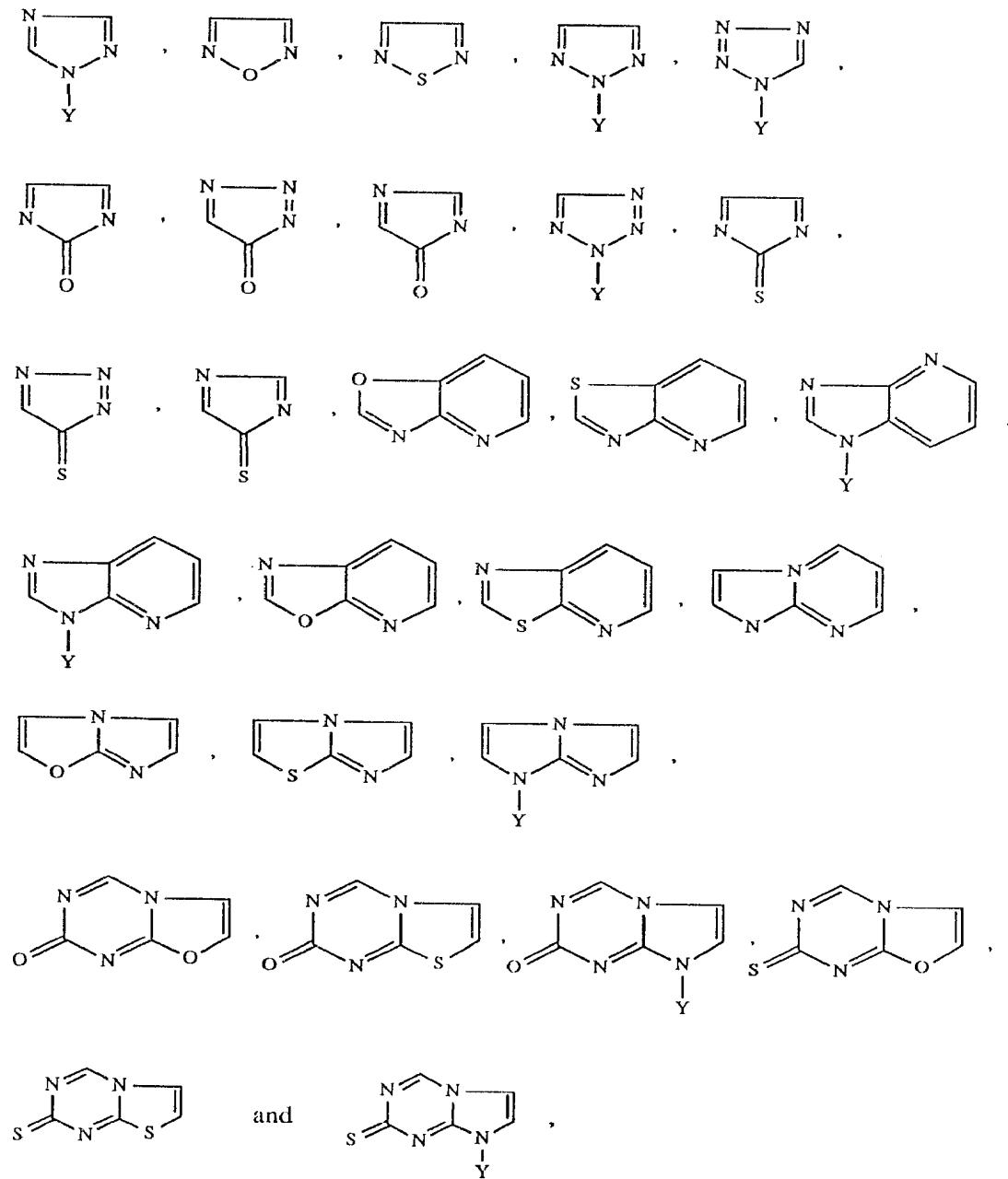
5. A compound according to claim 4 wherein the heterocyclic radical A contains from one to three hetero atoms from the group oxygen, sulfur and nitrogen, of which one hetero atom is always nitrogen, not more than one oxygen atom or sulfur atom being present.

6. A compound according to either claim 1 or claim 2 wherein the heterocyclic radical A is a heterocyclic basic structure, bonded via a carbon atom to the radical of the molecule of the compound of formula I, from the group





- 90 -



which basic structure is unsubstituted or, depending on the number of substituents possible in the ring system, can carry up to four of the substituents defined in claim 1, and wherein

E is C₁-C₃alkyl and Y is hydrogen, C₁-C₃alkyl or cyclopropyl.

7. A compound according to claim 6 wherein the heterocyclic radical A is unsubstituted or carries from one to three substituents from the group halogen, C₁-C₃alkyl, C₁-C₃haloalkyl and C₁-C₃haloalkoxy each having from 1 to 7 halogen atoms, and C₁-C₃alkoxy.

8. A compound according to claim 7 wherein the radical A is pyridyl or thiazolyl.

9. A compound according to either claim 1 or claim 2 wherein the radical B is a phenyl, pyridyl or thiazolyl radical each of which is unsubstituted or substituted by one or two radicals from the group halogen, C₁-C₃alkyl, C₁-C₃haloalkyl and C₁-C₃haloalkoxy each having from 1 to 7 halogen atoms, and C₁-C₃alkoxy.

10. A compound according to claim 7 wherein the radical A is 3-pyridyl, 2-halopyrid-5-yl, 2,3-dihalopyrid-5-yl, 2-halothiazol-4-yl, 1-oxopyrid-3-yl, 1-oxo-2-halopyrid-5-yl or 1-oxo-2,3-dihalopyrid-5-yl.

11. A compound according to either claim 1 or claim 2 wherein R₁ is hydrogen, R₂ is methyl, ethyl or cyclopropyl, and A is pyridyl, 1-oxopyridyl or thiazolyl, or pyridyl, 1-oxopyridyl or thiazolyl each of which is substituted by from one to three substituents from the group halogen, C₁-C₃alkyl, C₁-C₃haloalkyl and C₁-C₃haloalkoxy each having from 1 to 7 halogen atoms, and C₁-C₃alkoxy.

12. A compound according to either claim 1 or claim 2 wherein R₁ is hydrogen.

13. A compound according to claim 12 wherein R₁ is hydrogen and R₂ is methyl.

14. A compound according to claim 1 wherein R₃ is C₁-C₃alkyl, cyclopropyl, cyclohexyl, phenyl, benzyl or the radical -CH₂-COO-CH₃.

15. A compound according to claim 13 wherein R₃ is benzyl substituted by from 1 to 3 ring substituents from the group fluorine, chlorine, bromine, C₁-C₂alkyl, C₁-C₂haloalkyl, C₁-C₂alkoxy, C₁-C₂alkylthio, nitro and cyano.

16. A compound according to claim 13 wherein R₃ is phenyl substituted by from 1 to 3 ring substituents from the group fluorine, chlorine, bromine, C₁-C₂alkyl, C₁-C₂haloalkyl,

C₁-C₂alkoxy, C₁-C₂alkylthio, nitro and cyano.

17. A compound according to claim 13 wherein R₃ is C₁-C₆alkyl substituted by a hydroxy group.

18. A compound according to claim 13 wherein R₃ is C₁-C₆alkyl substituted by a C₁-C₅alkoxycarbonyl group.

19. A compound according to claim 13 wherein R₃ is -CH₂CH₂F, -CH₂CH₂Br, -CH₂CH₂CH₂Cl, -CH₂CH₂CH₂Br or -CH₂CHClCH₂CH₂Cl.

20. A compound according to claim 13 wherein R₃ is -CH₂CH₂O-CH₃, -CH₂CH₂CH₂-O-CH₂CH₃, -CH(CH₃)CH₂-O-CH₃, -CH₂CH(OCH₃)₂, -CH₂CH₂-N(CH₂CH₃)₂, -CH₂-CH₂CH₂-N(CH₃)₂ or -CH₂CH₂CH₂-N(CH₂CH₃)₂.

21. A compound according to claim 13 wherein R₃ is C₄-C₆cycloalkyl that is unsubstituted or substituted by one or two C₁-C₄alkyl radicals.

22. A compound according to claim 21 wherein R₃ is cyclopentyl or cyclohexyl.

23. A compound according to claim 21 wherein R₃ is C₃-C₆cycloalkyl substituted by one or two methyl groups.

24. A compound according to either claim 1 or claim 11 wherein A is 2-chlorothiazol-4-yl, 2,3-dichloropyrid-5-yl, 1-oxopyrid-3-yl or 1-oxo-2-chloropyrid-5-yl; R₂ is methyl and R₃ is cyclopropyl, -CH₂CH₂Cl, -CH₂CH(OCH₃)₂ or -CH₂CH₂N(CH₃)₂.

25. A compound according to claim 1 wherein A is 2-chlorothiazol-4-yl.

26. A compound according to claim 1 wherein A is 2-chloropyrid-5-yl.

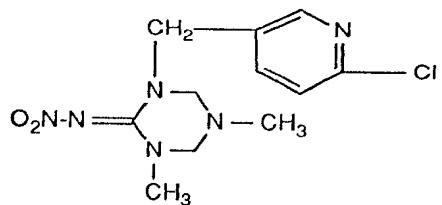
27. A compound according to claim 1 wherein A is 1-oxo-2-chloropyrid-5-yl or 1-oxopyrid-5-yl.

28. A compound according to claim 1 wherein A is 2-chloropyrid-5-yl, 2,3-dichloropyrid-5-yl, 2-chlorothiazol-4-yl, 1-oxopyrid-3-yl or 1-oxo-2-chloropyrid-5-yl;

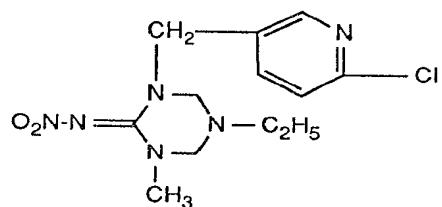
- 93 -

R₁ is hydrogen; R₂ is methyl; and R₃ is n-propyl.

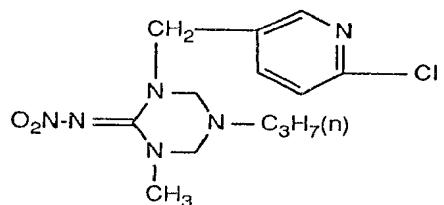
29. A compound according to claim 26 of the formula



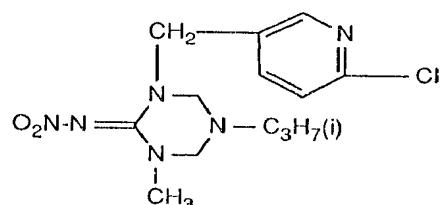
30. A compound according to claim 26 of the formula



31. A compound according to claim 26 of the formula

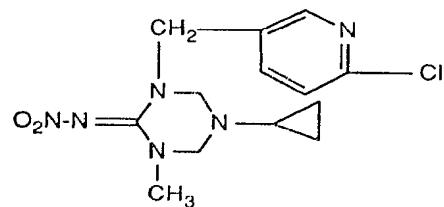


32. A compound according to claim 26 of the formula

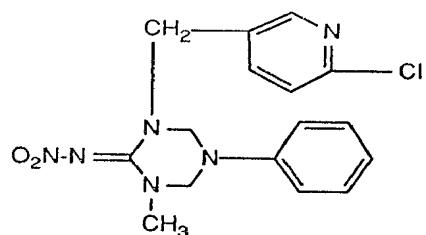


- 94 -

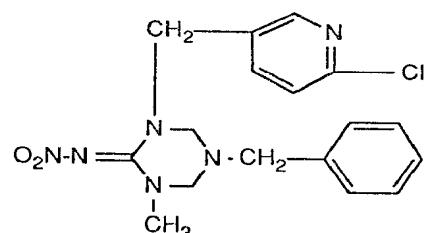
33. A compound according to claim 26 of the formula



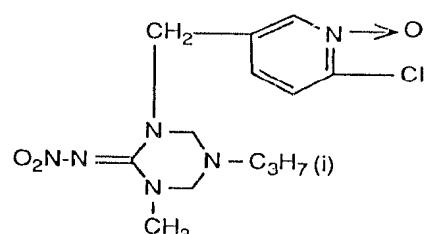
34. A compound according to claim 26 of the formula



35. A compound according to claim 26 of the formula

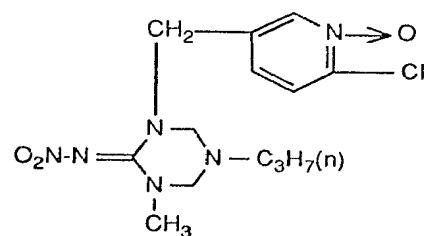


36. A compound according to claim 14 of the formula

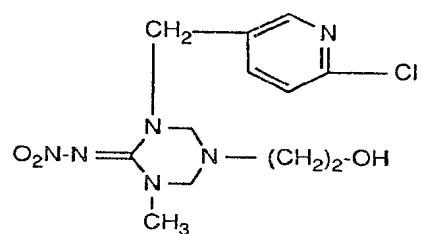


- 95 -

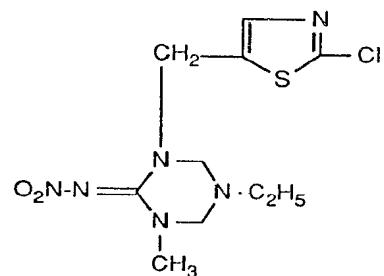
37. A compound according to claim 14 of the formula



38. A compound according to claim 26 of the formula

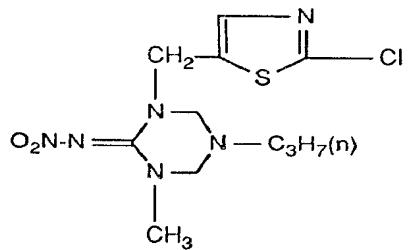


39. A compound according to claim 25 of the formula

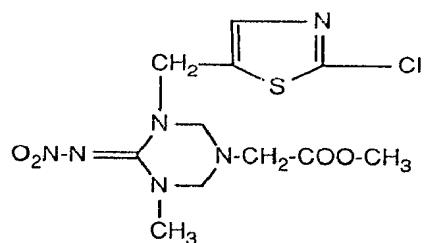


40. A compound according to claim 27 of the formula

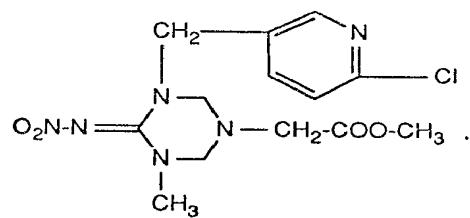
- 96 -



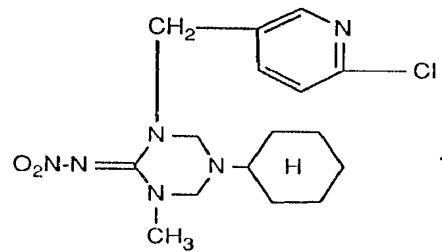
41. A compound according to claim 14 of the formula



42. A compound according to claim 14 of the formula

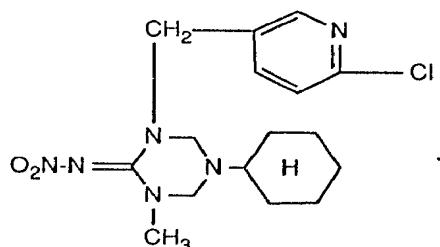


43. A compound according to claim 14 of the formula

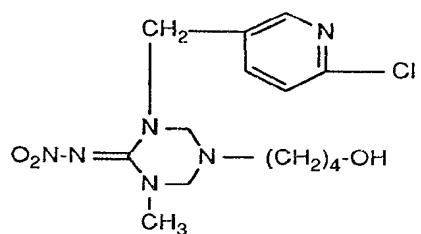


- 97 -

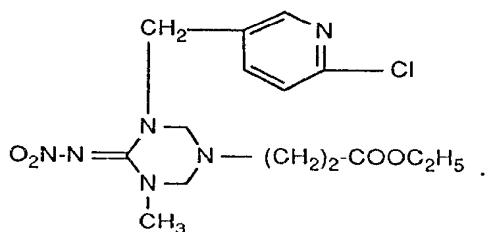
44. A compound according to claim 14 of the formula



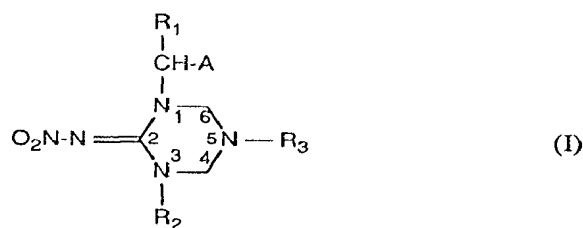
45. A compound according to claim 26 of the formula



46. A compound according to claim 26 of the formula



47. A process for the preparation of a compound of formula I according to claim 1



wherein

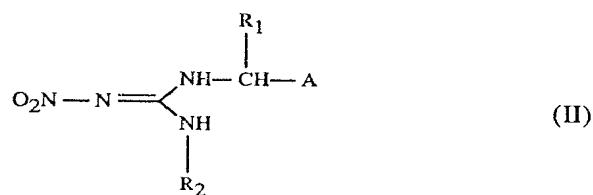
R_1 is hydrogen or $C_1\text{-}C_4$ alkyl;
 R_2 is hydrogen, $C_1\text{-}C_6$ alkyl, $C_3\text{-}C_6$ cycloalkyl or a radical $-\text{CH}_2\text{B}$;
 R_3 is hydrogen; $C_1\text{-}C_{10}$ alkyl; $C_3\text{-}C_6$ cycloalkyl; $C_1\text{-}C_{10}$ alkyl substituted by from 1 to 12 radicals from the group halogen, hydroxy, $C_1\text{-}C_4$ alkoxy, $C_1\text{-}C_4$ haloalkoxy having from 1 to 9 halogen atoms, di-($C_1\text{-}C_4$ alkyl)amino and $C_1\text{-}C_5$ alkoxycarbonyl; $C_3\text{-}C_6$ cycloalkyl substituted by from 1 to 4 $C_1\text{-}C_4$ alkyl radicals or halogen atoms; $C_2\text{-}C_8$ alkenyl or $C_2\text{-}C_8$ alkynyl; $C_2\text{-}C_8$ alkenyl or $C_2\text{-}C_8$ alkynyl each of which is substituted by from 1 to 6 halogen atoms; phenyl; benzyl; or phenyl or benzyl each of which is substituted by from 1 to 3 ring substituents from the group halogen, $C_1\text{-}C_4$ alkyl, $C_1\text{-}C_4$ haloalkyl having from 1 to 9 halogen atoms, $C_1\text{-}C_4$ alkoxy, $C_1\text{-}C_4$ haloalkoxy having from 1 to 9 halogen atoms, $C_1\text{-}C_4$ alkylthio, nitro and cyano;

- A is an unsubstituted or mono- to tetra-substituted aromatic or non-aromatic, monocyclic or bicyclic heterocyclic radical that can have one or two substituents from the group $C_1\text{-}C_3$ haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl having from 1 to 3 halogen atoms, $C_2\text{-}C_3$ alkenyl, $C_2\text{-}C_3$ alkynyl, $C_2\text{-}C_3$ haloalkenyl and $C_2\text{-}C_3$ haloalkynyl each having from 1 to 4 halogen atoms, $C_1\text{-}C_3$ haloalkoxy having from 1 to 7 halogen atoms, $C_1\text{-}C_3$ alkylthio, $C_1\text{-}C_3$ haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, and from one to four substituents from the group $C_1\text{-}C_3$ alkyl, $C_1\text{-}C_3$ alkoxy and halogen; and
- B is phenyl; cyanophenyl; nitrophenyl; halophenyl having from 1 to 3 halogen atoms; phenyl substituted by $C_1\text{-}C_3$ alkyl, $C_1\text{-}C_3$ haloalkyl having from 1 to 7 halogen atoms, $C_1\text{-}C_3$ alkoxy or by $C_1\text{-}C_3$ haloalkoxy having from 1 to 7 halogen atoms; 3-pyridyl; 5-thiazolyl; 5-thiazolyl substituted by one or two substituents from the group $C_1\text{-}C_3$ alkyl, $C_1\text{-}C_3$ haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, $C_2\text{-}C_3$ alkenyl, $C_2\text{-}C_3$ alkynyl, $C_1\text{-}C_3$ alkoxy, $C_2\text{-}C_3$ haloalkenyl and

C_2 - C_3 haloalkynyl each having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, halogen, cyano and nitro; or 3-pyridyl substituted by one or two radicals from the group C_1 - C_3 haloalkyl having from 1 to 7 halogen atoms, cyclopropyl, halocyclopropyl, C_2 - C_3 alkenyl, C_2 - C_3 alkynyl, C_2 - C_3 haloalkenyl and C_2 - C_3 -haloalkynyl having from 1 to 4 halogen atoms, C_1 - C_3 haloalkoxy having from 1 to 7 halogen atoms, C_1 - C_3 alkylthio, C_1 - C_3 haloalkylthio having from 1 to 7 halogen atoms, allyloxy, propargyloxy, allylthio, propargylthio, haloallyloxy, haloallylthio, cyano and nitro, or by from one to four radicals from the group C_1 - C_3 alkyl, C_1 - C_3 alkoxy and halogen;

or a salt thereof with an inorganic acid, which process comprises

- a) reacting a compound of formula II

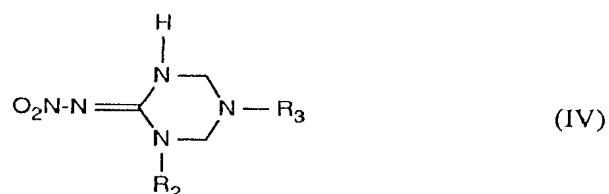


with formaldehyde, or paraformaldehyde, and a compound of formula III



or

- b) reacting a compound of formula IV



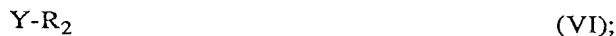
- 100 -

with a compound of formula V



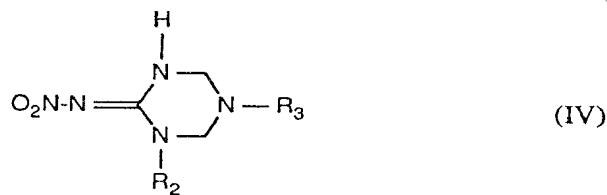
or

- c) for the preparation of a compound of formula I wherein R₂ is other than hydrogen, reacting a resulting compound of formula I wherein R₂ is hydrogen with a compound of formula VI



and, if desired, converting a resulting compound of formula I into a salt thereof; R₁, R₂, R₃ and A in formulae II to VI being as defined in claim 1, X being a halogen atom and Y being a leaving group.

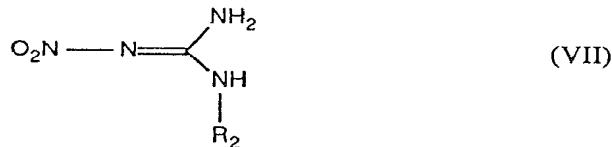
48. A compound of formula IV according to claim 47



wherein R₂ and R₃ are as defined in claim 1, with the exception of 2-nitroimino-5-methyl-1,3,5-triazacyclohexane and 2-nitroimino-1,3,5-triazacyclohexane.

49. A process for the preparation of a compound of formula IV according to claim 48, which comprises reacting a compound of formula VII

- 101 -



with formaldehyde, or paraformaldehyde, and a compound of formula III



R_2 and R_3 in formulae VII and III being as defined in claim 1.

50. A pesticidal composition comprising a compound according to claim 1 as active ingredient, together with suitable carriers and/or other adjuvants.

51. A method of controlling insects and representatives of the order Acarina, wherein the pests or their various development stages, or the locus thereof, are brought into contact or treated with a pesticidally effective amount of a compound of formula I according to claim 1 or with a composition comprising a pesticidally effective amount of such a compound together with adjuvants and carriers.

52. A method according to claim 51 for controlling plant-destructive insects.

53. A method according to claim 52 for controlling sucking insects.

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**Fetherstonhaugh & Co.,
Ottawa, Canada
Patent Agents**

SUBSTITUTE

REPLACEMENT

SECTION is not Present

Cette Section est Absente